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ORIGINAL ARTICLE

A Review of Crude Oil Recovery Methods from Petroleum Sludge

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ABSTRACT – The expansion of petrochemical refinery industries led to the exponential increase of petroleum sludge production and its disposal. Consequently, eliminating petroleum sludge from storage, processing, transportation, and pipeline systems presents a significant problem for the oil and gas industry. This is a challenge as mismanagement of petroleum sludge could mean the introduction of hazardous elements into the environment. After all, there are an estimated 60 million tons of petroleum sludge produced annually. According to statistics, one ton of sludge is generated for every 500 tons of crude oil extracted. Therefore, the petroleum industry requires a more effective and environmentally friendly techniques as an alternative treatment method for petroleum sludge. Currently, the most viable approach is oil recovery via solvent extraction. This method has been extensively used for petroleum sludge treatment under various experimental conditions. The reason is that solvent extraction is one of the cheapest recycling processes. It is also environmentally friendly to an extent as it uses lubricating oil that does not require water, and the solvent is reusable. However, due to its low efficiency and significant uncertainty in factors like solvent to sludge ratio and extraction time, solvent extraction alone is insufficient to recover crude oil from a large volume of petroleum sludge. Therefore, combining different solvent extraction methods and freeze-thaw hybrid techniques is proposed as a viable approach to improve oil extraction efficiency. In retrospect, this paper reviews thorough analysis of petroleum sludge's physical and chemical properties and its oil recovery using hybrids of different methods to promote the waste-to-wealth initiative.

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INTRODUCTION

Petroleum sludge is a solid emulsified waste produced by the oil and gas sector [1]. It contains various amounts of waste oil, wastewater, aliphatic and aromatic hydrocarbons, heteroatoms, and solid particles. All these components that make up the sludge threaten the destruction and balance of the ecosystem as it is toxic and harmful. Sludge must be treated and rendered harmless before being disposed of. The proper management and disposal of these wastes prevent the pollution of surface and groundwater, the contamination of the surrounding air, and the possibility of fires, explosions, food chain poisoning, and the loss of green areas [2]. In addition, there is a need to process sludge more effectively as crude oil is a major component of petroleum sludge and has a high recycling value.

As a result, more oil recovery technologies have been developed to treat and recycle petroleum sludge in recent years. Such oil recovery technologies are in high demand, and better treatment methods have been discovered and invented too; a prominent one being recycling. As it allows the petroleum industry to reuse valuable oil for reprocessing, reformulating, and energy recovery, recycling is the most environmentally responsible option for dealing with petroleum sludge [3]. Furthermore, recycling petroleum sludge reduces the amount of toxic waste disposed of outside of the industrial region, reduces pollution, and reduces the usage of non-renewable energy resources. To sum up, petroleum sludge can be use to minimize the amount of hazardous waste solids disposed of and the degree of contamination they cause, and the use of non-renewable resources [1].

One of the proposed treatment method for petroleum sludge is solvent extraction. It is a process that requires combining petroleum waste and solvent in an appropriate ratio to provide sufficient oil miscibility in the solvent, where most water and particles are discarded as undesirable contaminants can be extracted using gravity or centrifugation [4]. Previous solvent extraction research was primarily concerned with tank bottom sludge, with little attention paid to petroleum refinery wastewater treatment pond sludge, which contains more water and less oil than tank bottoms. However, in the solvent extraction process, such high-moisture sludge tends to create emulsified water during vigorous agitation, and the water cannot be successfully removed by subsequent gravity settling, resulting in high residual water content in the extractant [5]. As a result, the extractant may contain unwanted contaminants, thus decreasing the quality of the recovered oil. Furthermore, when recovered oil is reused as a fuel for further reprocessing, it can cause refining equipment corrosion and catalyst poisoning [6], [7].

Proper management of solid waste that is cost-effective and environmentally friendly is a significant concern by the industrial players due to the scale of petroleum extraction today. Therefore, petroleum waste management is required to

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manage waste from its inception to its final disposal, including waste collection, treatment, and disposal [2]. Compared to treatment and disposal, recycling is currently considered the most environmentally sustainable way of disposing and handling petroleum sludge due to environmental friendliness, rising disposal costs, and strict environmental policy [3]. In addition, it optimizes the use of petroleum sludge services and reduces the amount and degree of contamination generated by the sludge. Regardless, the current recycling methods of petroleum sludge are now deemed impractical due to treatment costs and environmental policies.

In general, most petroleum sludge treatment methods use three-phase waste management methods: minimize petroleum sludge output by technology, extract oil from petroleum sludge, and dispose of unrecoverable petroleum sludge [3]. The first phase of waste management aims to reduce petroleum sludge production. Meanwhile, the second and third phases focus on improving the quality of petroleum sludge treatment [4]. However, all the methods that have been developed have limitations and factors that affect the oil recovery rate. On account of this, this comprehensive review analyzes and concludes the optimum oil recovery method from petroleum sludge.

PETROLEUM SLUDGE SOURCE AND ITS FORMATION

Vast amounts of petroleum sludge can be generated during upstream operations such as crude oil extraction, transportation, storage, and downstream processes such as crude oil refining. Al-Futaisi *et al.* [8] categorized petroleum sludge as simple oil or sludge based on the proportion of water and particles in the oily matrix. In general, sludge has more water than waste oil, is more viscous, and has a larger percentage of particulates. A stable water-in-oil (W/O) emulsion is the most common physical type of oil sludge waste [9]. Upstream operations, slop oil from oil fields, crude oil tank bottom sediments, and drilling mud residues contribute to petroleum sludge formation. In addition, slop oil emulsion solids, heat exchange package cleaning sludge, and remains from oil/water separators such as the American Petroleum Institute (API) separator, parallel plate interceptor, and corrugated plate interceptor (CPI) all contribute to oil sludge in downstream processes. Sediments at the bottom of rail, truck, or storage tanks can also generate sludge downstream. For example, sludge from flocculation-flotation units (FFU), dissolved air flotation (DAF), induced air flotation (IAF) units, and excess activated sludge from on-site wastewater biological treatment [2]. When crude oil is temporarily stored in storage tanks, it tends to break into heavier and lighter petroleum hydrocarbons (PHCs), resulting in petroleum sludge. The petroleum sludge-like mixture would settle with heavier PHCs, solid particles, and water. It is then dumped in a storage tank [10], [11]. Figure 1 illustrates petroleum sludge originating from upstream, midstream, and downstream sources.

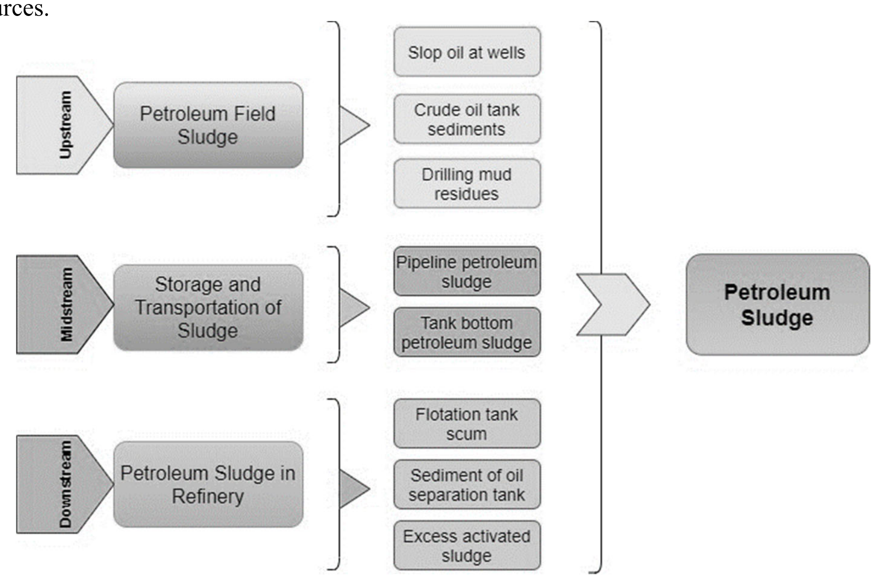

Figure 1. The sources of petroleum sludge [12].

COMPOSITION OF PETROLEUM SLUDGE

Mazlova & Meshcheryakov [13] described petroleum sludge as a recalcitrant residue, a stable water-oil emulsion of water solids, petroleum hydrocarbons (PHCs), and metals. The total petroleum hydrocarbon (TPH) content of petroleum sludge varies depending on the mass. Usually, the TPH content can range from 15-50%, with water contents at 30-85% and solids from 5-46% [14]. PHCs and organic compounds in petroleum sludge are divided into four fractions: saturates, aromatics, resins, and asphaltenes [15]. In average, it comprises 40-52% alkanes, 28-31% aromatics, 8-10% asphaltenes, and 7-22.4% of resins. Nitrogen (N) is often included in the distillate residue as part of the asphalt and resin fraction. Asphaltenes and resins, which contain hydrophilic functional groups that serve as lipophilic emulsifiers, stabilize the petroleum sludge emulsions.

Petroleum sludge comes from different sources in the refinery: separators, storage tanks bottoms, and floating sludge from industrial wastewater treatment plants. These residues contain carbonaceous solids, supernatant oil, free or emulsified water. The sludge composition is complex, including oil-in-water (O/W), water-in-oil (W/O), and suspended solids, usually characterized by stable emulsions of W/O, water, solids, PHC, and metals. The W/O emulsion has a high oil concentration, and its stability depends on a protective film that prevents water droplets from aggregating. This interfacial film comprises several natural emulsifiers, such as fine solids, PHC (asphaltenes and resins), oil-soluble organic acids, and other finely segregated products [14], [16], [17].

According to the American Petroleum Institute (API), the composition of petroleum sludge include both organic and heavy metal components in typical concentrations. The pH of petroleum sludge typically ranges between 6.5 and 7.5. Its composition varies from every refinery and from petroleum sludge to another, depending on the type of refined source, processing scheme, crude oil, and equipment [18]. Petroleum sludge contains various compounds and varies in composition. However, the basic components can be concluded to be a large amount of solids, water, and hydrocarbons, many of which are toxic, mutagenic, and carcinogenic, such as metallic chemical elements and Polycyclic Aromatic Hydrocarbons (PAHs) [19].

Other than the variation in the basic components, another factor that needs to be considered when designing petroleum sludge treatment methods is petroleum sludge's physical qualities (such as viscosity, density, and calorific value). The physical qualities can vary significantly due to its varied chemical compositions. The polarity and molecular weight of chemical species in the sludge can also considerably influence the physical characteristics of petroleum sludge [20]. There is also a diversity of heavy metals in the components of petroleum sludge, resulting from various sources, and their concentrations and species can change over a wide range, like organic compounds [14]. The metal concentration in petroleum sludge from various refineries is reported in various literature can affect the method of oil recovery.

PETROLEUM SLUDGE TOXICITY AND ENVIRONMENTAL IMPACT

The inappropriate disposal of petroleum sludge that results from the inefficient design of petroleum sludge treatment methods is harmful to the environment. It might pose a conspicuous long-term threat to our ecosystem due to the high quantities of dangerous chemicals. Furthermore, after entering the terrestrial environment, petroleum waste can interact with soils' physical and chemical properties resulting in soil morphological alteration [21]. Due to their high viscosity, petroleum sludge components can be fixed in soil pores, adsorbed the surface of soil mineral constituents, or create a continuous coating on the soil surface [22]. Consequently, soils' hygroscopic moisture, hydraulic conductivity, and water retention capacity would be decreased [22], [23]. In addition, some components in the sludge can linger on the soil's surface and form a crust that reduces water availability and restrict water/air exchange.

Nutrient deficiency and stunted growth in receiving soil vegetation have been reported by Al-Mutairi *et al.* [24]. Tang et al. [25] studied that higher molecular weight components in petroleum sludge and its degrading products may remain close to the soil's surface, form hydrophobic crusts, reduce water accessibility, and restrict water or air exchanges. Petroleum sludge can also alter the physical and chemical properties of the soil, resulting in morphological changes and soil destruction [26]. As a result, petroleum contains genotoxic hydrocarbons and PAHs to living creatures, which are a major concern to health because of inadequate treatment and improper disposal [21]. In addition, PHCs may penetrate the soil and enter underground water, posing a significant threat to marine life [22]. The petroleum sludge contains a high concentration of PHCs and PAHs. Most of these components are recalcitrant because of their tight molecular bonds, high molecular weight, hydrophobicity, and low water solubility.

The polycyclic aromatic hydrocarbons (PAHs), which are genetically toxic to humans and other living organisms, are an utmost root of concern. The petroleum hydrocarbon PHCs in the petroleum sludge could travel down to the soil and affect the groundwater connected to the aquatic systems, resulting in significant adverse outcomes such as decreased fish diversity and habitat. Besides that, after a long period in the environment, the weathered or aged chemical residues will resist desorption and degradation. Sludge can also be a source of atmospheric volatile organic compound (VOCs) pollution when deposited in landfills/lagoons. This can increase the risk of health problems in the communities and workers. Due to the danger of petroleum sludge, many countries have enacted stringent legislation for handling, storage, and disposals, such as the Resource Conservation and Recovery Act (RCRA) [14].

SIGNIFICANT OF PETROLEUM SLUDGE TREATMENT

The processing of sludge and the recovery of valuables are two key issues that oil companies face. Every year, an innovation in the petroleum industry emerges concerning the use of slop oil and paraffin waxes separated from crude oil during storage and pipeline transfers to reduce maintenance costs, corrosion, and environmental problems associated with sludge remediation. Like in other developed and developing countries, hazardous waste has become a significant environmental and public health issue in Malaysia. According to Malaysia's Environmental Quality (Scheduled Wastes) Regulations in 2005, the waste oil, also known as petroleum sludge, which is listed as code SW 310, should be appropriately managed. Waste oil with an economic value can be recovered by waste oil recovery facilities licensed by the Department of Environment. Even if the recovered, recycled, or reconstituted waste oil does not meet the standards and specifications, it is still classified as scheduled waste. Because of the hazardous nature of petroleum sludge, disposal of excess sludge will be prohibited soon, prompting increased interest in potential sludge treatment technology. Therefore, refiners have two primary goals for sludge treatment. The first goal is to improve the recovery of valuable oil from petroleum sludge, and the second goal is to find a way to dispose of treated sludge in an economical, fast, and environmentally friendly manner.

OVERVIEW OF PETROLEUM SLUDGE MANAGEMENT AND TREATMENTS

Waste management plays a vital role in achieving sustainable waste management following waste regulations. As a prominent type of solid waste, petroleum sludge requires a particular and effective treatment method. In re to limited on-site storage space, rising disposal costs, environmental concerns, and stringent legislation, the petroleum industry has devised a novel approach to treating and managing petroleum sludge [14].

Despite these efforts, several pressing issues have arisen regarding waste disposals, such as health and safety concerns and improper landfill management (Ministry of Housing and Local Government Malaysia, 2005). According to the Directorate-General of the Environment of the European Commission (2012), a plan needs to be developed to include possible waste management. The waste management hierarchy is illustrated in Figure 2. The hierarchy's goal is to separate the negative environmental impacts of resources and create a recycling system. An example of each element in the hierarchy is shown in Table 1.

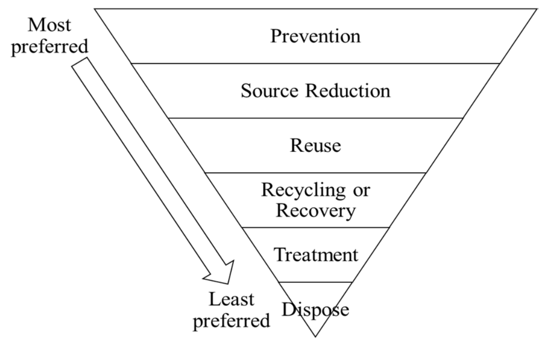

Figure 2. Hierarchy of waste management [27]

Table 1. Elements of waste management plan with their examples [28].

Waste management plan elements	Examples from the petroleum industry
Prevention	Separation of hazardous and non-hazardous wastes, develop new or modified procedures and operations to determine raw materials' water and sludge content.
Source reduction	The use of gravel packs and filters to reduce the amount of solids or sludge in the upstream segment area
Reuse	Due to the oil content, petroleum sludge is reused as part of the raw material in the treatment unit (such as coking)
Recycling or Recovery	The oil is collected from the bottom of the storage tank by centrifugation and filtration, the catalyst fines and petcoke are recycled, and the products of high value are extracted from the petroleum sludge using solvent extraction.
Treatment	Biological, chemical, thermal, physical processes
Dispose	Biodegradation, landfarming, incineration, composting, and landfilling

G. Hu *et al.* [14] claim that petroleum sludge management is a significant challenge for the petroleum industry. The best treatment and disposal methods for petroleum sludge are determined primarily by the sludge's physical and chemical properties, the availability of facilities, and regulatory requirements. Before final disposal, sludge is typically deoiled or dewatered (e.g., centrifuging). Following Hui *et al.* [1], the first step in sludge management is to prevent sludge formation, apply some techniques to reduce the amount of sludge formed, and then use effective treatment methods to recover the sludge.

It can be concluded that reduce waste and be more environmentally friendly, recovery and recycling are considered acceptable and sustainable methods for managing sludge. Oil recovery refers to the separation of oil from petroleum sludge, which is valuable for other uses in the petroleum industry. Like the other three R's of sustainability, recycling has proved to be one of the most effective ways to deal with petroleum sludge. Recycling can also help to reduce the amount of toxic petroleum sludge in storage tanks, avoiding emissions and lowering non-renewable energy usage.

Solvent Extraction

Solvent extraction is a method that mixes the petroleum sludge with extraction solvent at desired proportion to remove non-volatile and semi-volatile organic compounds [1], [3]. This method used the principle of solid-liquid two-phase extraction, which states that the solubility of various crude oil components in petroleum sludge varies depending on the extractants. It will break down the sludge's complex molecules into their simple constituents [2]. Generally, the type and quantity of extractant used in this process are critical, as different types and amounts result in different oil recovery rates. The solvent can be reused to complete the extraction process, and the solvent with impurities will be isolated and sent to a solvent recycling tank. Solvent extraction is a quick and effective technique in general. This technology can efficiently minimize the volume of petroleum sludge by separating petroleum sludge and solid residue in a short amount of time.

Freeze-Thaw

In freeze-thaw method, the water-phase and oil-phase separation can be split into two conditions [3]. The first condition is when the freezing point of hydrocarbons is lower than the water. When this condition happens, the petroleum's sludge water phase will be cooled and frozen first, and its volume will expand. With the continued decrease in temperature, the equilibrium of the two phases in the emulsion mixture will be steadily broken, and the oil phase will begin to coagulate [4]. The dual effects of oil-phase gravity and surface tension will progressively stratify the oil-water emulsion with the water phase during the subsequent thawing period, resulting in the oil-water emulsion's instability and, eventually, the separation and recovery of crude oil. The second situation is when the freezing point of hydrocarbons is higher than the water. The oil phase freezes first, followed by the water phase. The water phase will begin to freeze as the temperature drops, causing its volume to rise and the reservoir to burst. The oil and water phases can eventually stratify and detach due to gravity during the thawing process, after which the oil phase can be recycled [1].

Pyrolisis

Petroleum sludge is extracted using this process, which involves high-temperature pyrolysis and gasification of the organic components in the sludge with indirect heat transfer under anaerobic conditions. This method involves water evaporation, vaporization of light organic components, and the cracking decomposition of coke and other inorganic materials. Pyrolysis of petroleum sludge produces lower NO_x and SO_x emissions than incineration, and it also allows heavy metals in petroleum sludge to be concentrated in the final solid product [14]. However, various factors, such as temperature, heating rate, sludge characteristics, and chemical additives can restrict pyrolysis. Furthermore, the product may contain high PAH levels and is not cost-efficient [3].

Surfactant Enhanced Oil Recovery (EOR)

The surfactant treatment is based on the chemical cleaning theory, which uses the surfactant's hydrophilic and hydrophobic groups. The most common form of surfactant is an amphiphilic compound [14], which can reduce surface tension or interfacial tension between various types of liquids and between liquids and solids, allowing them to be more effective at removing organic contaminants [3]. Chemical surfactants are fast and inexpensive to use, but they are toxic to the environment and resistant to biodegradation, which raises concerns.

Centrifugation

In this process, a pre-treated petroleum sludge is drawn through high-speed spinning equipment with heavy centrifugal forces to break up its elements into various densities quickly. This is done to lower the viscosity and to improve centrifugation efficiency while conserving energy [3]. Centrifugation is an innovative, safe, and effective process for petroleum sludge treatment. It does not necessitate a large amount of energy. However, the plant's construction necessitates a large amount of space, is expensive, and poses an environmental risk. Besides that, centrifugation can cause noise problems [14].

Froth Flotation

Froth flotation captured tiny solids/oil droplets by using air bubbles in an aqueous slurry, and it is then floated and accumulated in a froth layer [3]. A specific amount of water and air is injected into the sludge, so the water film between oil droplets will break. This will help the oil droplet drift with air bubbles and float to the water's surface. However, when treating petroleum sludge with low moisture and high viscosity, a large amount of water is needed, and the sludge must be pre-treated to minimize viscosity and remove coarse solid particles [14]. Overall, this method is effective but can be affected by many factors, especially the sludge's properties.

Microwave Irradiation

Microwave frequency is used in this process. The microwave energy will result in rapid and efficient heating of the petroleum sludge. This process will cause demulsification of the mixture of oil and reduce viscosity and the breakdown of the heavier into lighter hydrocarbons. The performance of this method has a few limiting factors such as power, duration, and sludge's properties. Nevertheless, it can be considered a highly energy-efficient method as it can increase the energy of molecules in a short time. However, because of the equipment requirements and high operating costs, its implementation at the industrial scale is limited [3].

Electrokinetic

This method uses low-intensity direct current (DC) across a pair of electrodes, resulting in the electron's movement from the lower to higher concentration region. Despite its effectiveness, the electrokinetic method's efficiency can be harmed by various factors. Compared to other recovery methods such as centrifugation and pyrolysis, the electrokinetic process for oil recovery from petroleum sludge uses less energy. This method has only been done at the laboratory level, and further investigation will need to be done at a large-scale level [3].

Ultrasonic Irradiation

Ultrasonic waves are used to create compression and reflections in the treatment chamber. Lowering the solubility of water-oil mixtures is an efficient treatment method for separating solid-liquid in high concentration suspensions. The compression cycle applies positive pressure to the medium, while the rarefaction cycle applies negative pressure, causing microbubbles to form. Cavitation causes a rise in the temperature of the emulsion system and a decrease in its viscosity, which improves the mass transfer of the liquid process and causes the water-oil mixture to destabilize. This process is a highly effective method that does not pollute the atmosphere and can be completed in a limited amount of time. However, there are some limitations, including sonification strength and intensity, salinity, and the initial concentration of PHC [3]. Table 2 summarizes the advantages and disadvantages of current oil recovery methods.

Table 2. The oil recovery methods and their advantages and disadvantages [3].

Oil Recovery Methods	Advantages	Disadvantages
Solvent Extraction	a) Very simple and cheap b) High efficiency c) Good for small scale	a) Long extraction time b) Energy intensive for solvent recovery c) Require large amount of solvent which is toxic and highly flammable
Freezing & Thawing	a) Simple and easy to use b) Suitable for cold region for natural freezing c) Fast and short-term treatment	a) Low efficiency b) High energy consumption and cost when freezing
Microwave Radiation	a) Environmentally friendly b) Reduced solvent usage c) More economical d) Shorter extraction time e) Improved extraction yield	a) Low efficiency with certain target compounds b) Need filtration and centrifugation to remove solid residue c) Limit application industrial scale
Ultrasonic Radiation	a) Shorter extraction time b) Less solvent consumption c) Good viscosity effect d) No secondary pollution	a) High power consumption b) High cost of use and maintenance. c) Difficult to scale up
Surfactant Enhanced Oil Recovery	a) High dehydration property b) Strong operability c) fast and cost-effective	a) Resistance to biodegradation b) Contain environmental toxicity.
Sludge Pyrolysis	a) Can recover oil in liquid phase b) Convenient for storage and transportation c) Produce less harmful waste gas d) Similar quality and property with low-grade petroleum distillates from refinery e) can use directly to diesel engine	a) Not cost-effective b) contain high concentration of polycyclic aromatic hydrocarbons c) limit by temperature, chemical additives, and characteristics of sludge
Electromagnetic	a) Require less energy b) Fast and efficient	a) Still in laboratory level b) Complicated application and in small scale
Froth Flotation	a) Can control the property of sludge with flotation reagent b) Can separate any mineral c) Simple application and low energy consumption	a) Costly b) Inconvenience in operation c) Low efficiency
Centrifugation Treatment	a) Low energy consumption b) Clean and efficient	a) Require large space for installation b) Very costly c) Pose environmental concern with noise and pollution

OIL RECOVERY FROM PETROLEUM SLUDGE

Due to the high oil concentration in the sludge, oil recovery from the petroleum sludge is considered one of the possible environmental options. In addition, pollution can be reduced, and hazardous waste can be disposed of. A comparative analysis based on other published reports on the most effective and simple method for treating petroleum sludge proved that the solvent extraction process is highly recommended. Table 3 summarizes all the methods for oil recovery from petroleum sludge. It is proven that the most used method is solvent extraction with maximum oil recovery that varies between 80% to 97% depending on the solvent used for extraction. The minimum oil recovery is a solvent extraction method that uses toluene and methyl ethyl ketone (MEK) as solvents with 37% and 30% recovery. Furthermore, hexane is the best solvent that gives the highest recovery, with 68% recovery. Hexane is the most used solvent for extracting oils due to its low boiling temperature, easy recovery from extractants, and soluble in hexane petroleum sludge.

Table 3. Overview of literature review of crude oil extraction method from oil sludge.

Origin of the petroleum sludge	Methods used to extract crude oil	Oil recovery (%)	Authors
New Jersey, USA	Dehydration and solvent extraction	55%	[29]
Niger delta area of Nigeria	Solvent extraction by using: i. Hexane	Hexane: 67.48% Kerosene: 62.55%	[30]

	ii. Kerosene		
United Arab Emirates	Solvent extraction by using MEK and LPG condensate	MEK: 39% LPG: 32%	[31]
Suez, Egypt	Solvent extraction by using heptane, toluene, kerosene, naphtha, methylene dichloride, ethylene dichloride, and diethyl ether	Varied between 80% to 97% depending on the solvent used for extraction	[32]
Azzawiya oil refinery in Libya	Solvent extraction by using dichloromethane (DCM)	42%	[33]
Niger Delta of Nigeria	Single solvent and combination (solvent blend)	66.25%	[34]
Western Canada	Combination method of solvent extraction and freeze-thaw by using different solvents: i. Cyclohexane (CHX) ii. Methyl ethyl ketone (MEK) iii. Ethyl acetate (EA)	62%	[34]
Kurdistan, Iraq Sample of the area: i. Guwayer ii. Khurmala	Solvent extraction by using toluene	Guwayer: 68.5% Khurmala: 75 %	[35]
Canada	Solvent extraction using MEK	62.2%	[36]
Tehran, Iran	Solvent extraction test with toluene and MEK	Toluene: 37.24% MEK: 30.41%	[37]
PETROBAS Northeast, Brazil	Solvent Extraction	13-53%	[38]
TK-C2/TK-C15	Solvent Extraction	75.94%	[32]
Taoyuan, Taiwan	Freeze Thaw	50%	[39]
Western Canada	Ultra-Sonic Irradiation	80%	[40]
Isthmus, Mexican	Solvent Extraction	16-55%	[41]
Manhattan, KS.	Solvent Extraction	67.5%	[30]
Mumbai, India	Froth Flotation	55%	[42]
Shengli Oilfield, China	Ultra-Sonic Irradiation	55.6%	[43]
Kerman, Iran	<u>Mechanical Shaking</u> i. Acetone ii. Chloroform iii. Dichloromethane iv. Hexane v. All 4 solvents <u>Mixer</u> i. Acetone ii. Chloroform iii. Dichloromethane iv. Hexane v. All 4 solvents <u>Solvent Extraction (Soxhlet/Reflux)</u> i. Acetone ii. Chloroform iii. Dichloromethane iv. Hexane v. All 4 solvents	i. 67.61% ii. 74.78% iii. 68.43% iv. 70.78% v. 73.76% i. 58.04% ii. 74.80% iii. 57.86% iv. 65.01% v. 73.81% <u>Number of Hydrocarbons Extracted</u> i. 108 ii. 78 iii. 79 iv. 206 v. 230	[44]
Baiji, Iraq	<u>Solvent Extraction</u> i. Methyl Ethyl Ketone	i. 95%	[45]
Kharg, Iran	<u>Solvent Extraction</u> i. Toluene ii. Methyl Ethyl Ketone	i. 37.24% ii. 30.41%	[37]
Wuhan, Hubei	<u>Solvent Extraction</u> i. Trichloromethane	i. 58.18%	[46]
Western Canada	<u>Solvent Extraction</u> i. Cyclohexane ii. Dichloromethane iii. Methyl Ethyl Ketone	i. 42.4 ± 0.8% ii. 37.7 ± 0.8% iii. 37.2 ± 1.9%	[4]

	iv. Ethyl Acetate v. 2-propanol Solvent Extraction & Freeze and Thaw i. Methyl Ethyl Ketone ii. Ethyl Acetate	iv. 37.6 ± 1.0% v. 17.1 ± 1.1% i. 62.6 ± 2.6% ii. 58.8 ± 2.8%	
-	Solvent Extraction i. n-Heptane ii. Ethylene Dichloride iii. Toluene iv. Methylene Dichloride v. Diethyl Ether vi. Naphtha cut vii. Kerosene cut	 i. 55.59% ii. 67.85% iii. 69.20% iv. 69.68% v. 59.99% vi. 78.03% vii. 80.97%	[32]
Arabian Gulf, UAE	Solvent Extraction i. Methyl Ethyl Ketone ii. LPG Condensate	 i. 39% ii. 32%	[47]
Niger delta area, Nigeria	Solvent Extraction i. Hexane ii. Kerosene	 i. 67.48% ii. 62.55%	[30]
Isthmus, Mexico	Solvent Extraction i. Supercritical Ethane	 i. 58.5 mass %	[41]

FACTORS THAT INFLUENCE OIL RECOVERY USING SOLVENT EXTRACTION

Numerous factors could affect the efficiency of oil sludge recovery using the solvent extraction method, including solvent types, solvent-to-sludge ratio, time of extraction, and solvent dose on the oil content.

Influence of Solvent Types

Various research has been published on using different solvents to extract oil from petroleum sludge. Gazinue *et al.* [38] extracted petroleum sludge using a Soxhlet extraction apparatus with supercritical ethane and dichloromethane. They discovered that removing more than half of the oil is possible. Taiwo and Otolorin [30] used hexane and xylene as solvents to extract petroleum hydrocarbons (PHCs) from petroleum sludge, with hexane having the maximum oil recovery rate of 67.5 percent. Abouelnasr and Zubaidy [31] reported that methyl ethyl ketone (MEK) and light petroleum gas condensate (LPGC) could extract 39% and 32% by mass of the initial sludge as recoverable oil, respectively. As the amount of solvent used increased, so did the grade of recovered oil in terms of solids and asphaltene content. The comparison study on other solvents is shown in Table 4.

Table 4. Comparison study on four different solvents using solvent extraction for petroleum sludge recovery.

Solvent	Characteristics	Advantages	Disadvantages
Cyclohexane	Alicyclic hydrocarbon with a six-carbon ring, colorless and clear liquid that has detergent-like odor.	Used as a recrystallization solvent. Many organic compounds exhibit good solubility in hot cyclohexane [48].	Flammable liquid. Poor solubility at low temperature [48].
Dichloromethane	Organochloride compound with colorless, volatile liquid, chloroform-like, sweet odor and is widely used as a solvent.	Able to dissolve a wide range of organic compounds. Extraction yields a light oil and potentially used as good feedstock oil [14].	Carcinogenic. Viscosity of crude oil decreases significantly with increase in temperature because of temperature on chemical structure of the ingredient of the crude oil [14].
Methyl Ethyl Ketone	Colorless organic liquid with an acetone-like odor.	Recovered oil by MEK exhibits an improved ash, carbon residue and asphaltene levels [31].	Recovered oil still contains high levels of sulfur and carbon residue thus requires purification before use as a fuel [31].
Ethyl Acetate	Organic compound with colorless liquid and has a characteristic sweet smell.	Volatile, relatively non-toxic, and non-hygroscopic. Can dissolve up to 3% water and has a solubility of 8% in water at room temperature [49].	Highly flammable. Unstable in the presence of strong aqueous bases and acids [49].

Influence of Solvent-to-Sludge Ratio

Generally, the oil recovery rate of solvent will increase with the solvent-to-sludge ratio until equilibrium is reached, with a ratio of 1:1 producing the lowest oil recovery rate [4]. This is because raising the solvent-to-sludge ratio can enhance the solubility of oil in solvent and consequently the amount of oil recovered. According to Hu *et al.* [4], the solvent-to-sludge ratio for the four solvents (cyclohexane, dichloromethane, methyl ethyl ketone, and ethyl acetate) was increased from 1:1 to 4:1, but there were no additional increases when it was higher than 4:1. Zubaidy & Abouelnasr [31] also investigated various ratios and discovered that a solvent-to-sludge ratio of 4:1 facilitated the extraction of the greatest amount of oil. An increase in this ratio exceeds 4:1, resulting in a slight increase in solvent recovery [4]. Therefore, the solvent-to-sludge ratio was determined to be the optimum ratio by the associated increased oil and solvent recovery and by the increased waste reductions.

Influence of Extraction Time

Extraction time is another critical component in solvent extraction performance because a long enough duration allows the solvent to dissolve the oil and allow contaminants to congregate to be separated by removal from the liquid phase [4]. Hu *et al.* found that 30 minutes was the optimal extraction duration, giving the required miscibility between petroleum sludge and solvent [4].

Influence of Solvent Dose on The Oil Content

As the solvent dose increases, so does the amount of oil that can be dissolved [31]. This trend is the same as Al-Doury [45]. Furthermore, the oil recovery rate will vary depending on the solvent used in the experiment, especially the solvent extraction method. Ahmad *et al.* [50] investigated the efficiency of hexane, pentane, benzene, heptane, and ether in recovering oil from refinery petroleum sludge while accounting for the impact of the most significant extraction method parameters. They discovered that heptane is the perfect solvent for solids separation. Naggar *et al.* [32] also described the same trend of increment on oil recovery rate when the solvent volume increases.

CONCLUSION

Since petroleum sludge is classified as a hazardous waste in many countries, inappropriate handling can harm the environment and human health. Considering the high oil content of sludge, oil recovery may be a preferable method since it may create profit while simultaneously reducing waste volume and pollutant concentration. As a result of increasingly rigorous rules prohibiting the direct land dumping of petroleum sludge, different physical, chemical, and biological approaches for its treatment have been developed. Comparing the various methods of petroleum sludge treatment, the solvent extraction method is the most often used treatment method for petroleum sludge. The solvent extraction method is the most straightforward and efficient approach for recovering petroleum sludge. Even though many studies have been done on petroleum sludge in the past, there is not much data on sludge properties or pre-treatment methods. As a result, this review study is essential in providing thorough literature on the subject.

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