Review:

An Overview of the Current State and Prospects for Oil Recovery from Oily Sludge

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Abstract: *Oily sludge is a solid emulsified waste created by the petroleum industry. Solid particles, crude oil, and water comprise most of their composition. Because it contains high concentrations of cycloalkanes, benzene series, polycyclic aromatic hydrocarbons, and other harmful and hazardous substances, it poses a severe risk to human health and the environment. It must be treated to reduce its toxicity. However, crude oil is a significant component of oily sludge and has a high recycling value. As a result, numerous procedures for extracting crude oil from oily sludge have been developed, including solvent extraction, pyrolysis, centrifugation, ultrasonic treatment, electronic treatment, flotation, supercritical treatment, and combination processes. The primary purpose of this review is to describe the evolution of various recycling technologies and to compare their benefits, drawbacks, and ways of action. This concept is expected to be the cornerstone for future recycling technology development.*

Keywords: *oily sludge; petroleum industry; hazardous; crude oil; toxicity*

INTRODUCTION

Energy is increasingly in demand globally because of the fourth industrial revolution's expanding economy. Most of the world's energy demands are met by fossil fuels, especially oil. With the accelerated expansion of industry and energy, consumption and demand for oil are rising drastically. As a result, refineries generate more oil sludge, which collects at the bottom of tanks when oil is being stored. This sludge negatively affects the tank's capacity and operational safety [1-5]. When discussing the production, exploration, storage, transportation, and processing of crude oil, the term "oil sludge" refers to a thick, viscous, and challenging-to-handle fluid [6-10].

A complex physicochemical combination of oil products, water, and mechanical contaminants, including clay, sand, and metal oxides, make up oil sludge. Oil sludge is mainly developed during producing, processing, storing, and transporting of oil. It comprises drilling fluid, waste oil in the well, emulsified solids produced during crude oil processing, and sediment found in the storage tank [11-14]. When oil is cleaned of pollutants and water, for example, oil sludge can occur as a consequence of natural, regulated processes, as well as other kinds of accidents (spills). The environment might sustain severe harm in the latter scenario due to a large-scale disaster or late identification. The primary goal of this review is to describe the evolution of various recycling technologies and compare their advantages, disadvantages, and methods of operation.

FORMATION OF OIL SLUDGE

During transportation to the refinery, the heavier and lighter crude oil hydrocarbons often separate. This problem is usually exacerbated by storing crude at low temperatures, removing volatile components, and using cool temperatures. The thick fractions that separate from crude oil and settle at the bottom of storage tanks are referred to as sludge [1,15]. Both upstream and downstream oil industry operations can generate large volumes of oil waste. While downstream activities refer to crude oil refining, upstream operations refer to crude oil extraction, transportation, and storage. Oil sludge sources in upstream operations include slop oil in oil wells, oil left in tank bottoms, and drilling mud deposits.

Crude oil is temporarily stored in storage tanks before being converted into petroleum products, where it usually separates into heavier and lighter petroleum hydrocarbons. Heavy hydrocarbons frequently sink with water and other particulates. Oil sludge is a substance that collects at the bottom of storage tanks and combines oil, sediments, and water. These are taken out through tank cleaning procedures and shipped for additional processing or disposal [16-17].

The amount of sludge created during the refining process is determined by various factors, including the crude oil's properties (such as density and viscosity), the way the refinery processes the crude, how oil is stored, and–most importantly–the capacity of the refinery [18-24]. The production of more oil sludge is typically correlated with cleaning higher power. One ton of oil sludge is generated for every 500 tons of crude oil treated, according to estimations [25-27]. According to assessments, more than 60 million tons of oily sludge can produce each year, and more than 1 billion tons of oily sludge have accumulated as a whole [28-30]. Fig. 1 shows the schematic diagram of the oily sludge sources. Furthermore, it is predicted that the total output of oily sludges will rise in response to the rising global demand for refined goods [29,31].

According to the physicochemical composition and method of formation, oil sludge is divided into five groups [32-34]:

(i) Natural oil sludge - waste generated at the bottom of various water bodies after an oil spill.

(ii) Oil sludge - waste generated during well drilling, various drilling fluids.

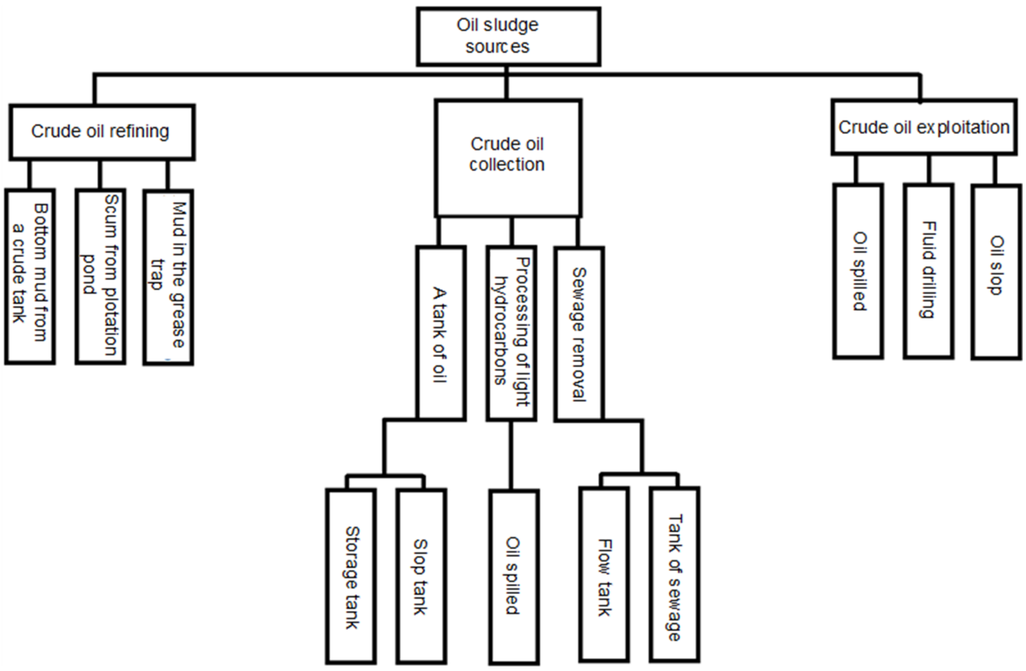

Fig 1. Schematic diagram of the oily sludge source

(iii) Oil sludge - waste produced during the purification of oil from solid hydrocarbons and mechanical impurities.
(iv) Tank oil sludge is waste generated throughout the transportation and storage of oil in various tanks.
(v) Ground oil sludge - is a product of the combination of soil and oil spilled on it; the reason for this can be both a technological process and an accident.

According to the state of aggregation, oil sludge is divided into liquid, solid and highly viscous, gaseous

■ CHARACTERISTICS OF OILY SLUDGE

Depending on its origin, oil sludge can have a vast array of constituents. The hydrocarbon component of oil waste comprises a variety of compounds that can be transformed into other compounds during long-term storage via condensation, isomerization, and polymerization. However, all oil sludge contains water and other impurities. Long-term storage of oil sludge changes over time due to precipitation accumulation, the advancement of microorganisms, the incidence of oxidative processes, and other processes. In some cases, oil sludge is a stable emulsion that cannot be separated into various components [35-36]. Oil sludge consists of organic constituents (72%), moisture (19.2%), sulfur (1.8%), and mineral part (16%). According to Alexandrovich and Sergeevich [37], the mineral composition of oil sludge was SiO_2: 4.55%, CaO: 3.14%, Fe_2O_3: 1.65%, Al_2O_3: 2.36%, MgO: 2.36%, and others: 3.3%.

Both inorganic and organic pollutants can be found in petroleum sludge. Metal compounds, including zinc, copper, lead, chromium, nickel, and mercury, are examples of inorganic pollutants. Total petroleum hydrocarbon measurements of organic pollutants in the sludge vary from 510,000 to 640,000 mg/kg [38]. Oil sludge has 15 to 50% petroleum hydrocarbons (in percent by weight), 30 to 85% water, and 5 to 46% solids. Petroleum hydrocarbons (PHCs) and further organic compounds in oily sludge are categorized into four types: Asphaltenes, aliphatic and aromatic chemicals, and compounds comprising nitrogen, sulfur, and oxygen (NSO) [39].

Alkanes, cycloalkanes, benzene, toluene, xylenes, naphthalene, phenols, and other polycyclic aromatic hydrocarbons, for instance, methylation derivatives of fluorine, phenanthrene, anthracene, chrysene, and pyrene account for up to 75% of PHCs in oily sludge [17]. Polar molecules such as naphthenic acids, mercaptans, thiophenes, and pyridines are noticed in the NSO fraction. The quantity of nitrogen (N) in oily sludge is less than 3%, with the bulk found in the distillate residue as a component of the asphalt and resin fraction [17].

In contrast to the oxygen concentration, typically less than 4.8%, sulfur (S) level can range from 0.3 to 10%. Asphaltenes are mixes of colloidal and pentane-insoluble chemicals that contain polyaromatic and alicyclic molecules with alkyl replacements (often methyl groups), and their molecular weights range from 500 to several thousand [40]. Since these components include hydrophilic functional groups and can thus act as lipophilic emulsifiers, asphaltenes and resins can be accountable for the oily sludge emulsion's stability. Oily sludge typically contains 40 to 52% alkanes, 28 to 31% aromatics, 8 to 10% asphaltenes, and 7 to 22.4% resins by mass [41]. The pH of oil sludge is typically between 6.5 and 7.5.

A recent study has identified significant amounts of Fe (60, 200 mg/kg), Zn (1.299 mg/kg), Cu (500 mg/kg), Ni (480 mg/kg), Cr (480 mg/kg), and Pb (565 mg/kg) in oil refinery sludges [41-44]. Additionally, oil sludge contains polycyclic aromatic hydrocarbons (PAHs), which are hazardous and cancer-causing-mutagenic, at amounts of 550 mg/kg or less.

The US Environmental Protection Agency (USEPA) has reported 16 PAHs as priority pollutants, namely: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoroanthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene, and benzo[g,h,i] perylene [45-46].

■ DANGER OF OIL SLUDGE TO THE ENVIRONMENT

Oily sludge is considered hazardous waste. If this is not handled and disposed of appropriately, it will have significant harmful effects. There are three possible ways that adverse consequences could manifest themselves. (1) Inadequately treated oily sludge pollutes surface waters, even groundwaters, and causes a severe excess of COD and oily substances in water; (2) The volatilization of oil components in oil sludge will result in an excessive concentration of total hydrocarbons in the air of nearby areas; and (3) Oily sludge comprises of an actual amount of toxic and harmful organic compounds, for instance, hydrocarbons, phenols, anthracene, and benzene ring compounds. As a result, oil sludge is classified on the National Inventory of Hazardous Wastes [36,47].

When oil sludge is subjected to environmental factors, it can expand, evaporate, be absorbed by living organisms, and undergo transformation [35]. The processes of oil-containing compound degradation are markedly accelerated by sunlight. In contrast, the spread of oil-derived products in the soil is markedly slowed down by the evaporation of light fractions. Heavy oil fractions eventually produce emulsions that are difficult to separate. The exposure temperature affects how quickly petroleum products decompose. Lower temperatures slow down the rate of decomposition. Therefore, bacterial, chemical, and photochemical breakdown processes, as well as some organisms' and plants' activities, all contribute to the degradation of petroleum products [48].

PHCs and other flame-retardant substances are present in large amounts. Oil sludge is categorized as hazardous waste in various countries; hence, incorrect removal or poor treatment can damage the environment and people's health. Oil sludge is categorized as a material of the third class of hazards or one with a medium environmental impact. The direct damage that oil sludge may do to the environment and humans is [49]:

(i) Chemicals from the first and second classes of hazards can be found in oil slime. Some examples include phenanthrenes, benzopyrene, and other pyrene derivatives. These compounds cause malignant tumors in animals, similar to those in people.

(ii) It damages the liver, heart, and brain. The oil contains harmful heavy metal ions that can enter human food systems and cause harm.

(iii) They cause slow seed germination and germination rate. According to laboratory tests, the seeds are entirely incapable of germination at a concentration of oil sludge of 6–8%. Because of their smaller size and underdeveloped root systems, the above-ground sections of plants in polluted soils seem weaker.

(iv) Animals that reside in soil polluted with oil waste, such as moles and mice, are doomed. Several oil compounds accumulate in the organisms of animals and have mutagenic effects, causing severe diseases, infertility, and affecting behavior.

■ OVERVIEW OF OIL SLUDGE DISPOSAL METHODS

Proper handling of oil waste is receiving more consideration due to expanded production and its hazardous nature. Numerous ways have been developed to control it to lower the concentration of harmful pollutants or immobilize them, limiting the impact of unfriendly material on the environment and health [50]. Among them are farming/burial, photocatalysis, incineration, curing/stabilization, sonication, solvent extraction, chemical processing, pyrolysis, and biodegradation [51-56].

The three waste management phases are used in most remediation methods: (1) reducing oil sludge creation through technology, (2) recovering oil from oily sludge, and (3) disposing of non-recoverable oil sludge [50]. Various techniques for treating oily sludge have been developed, as detailed below.

Because of the dangerous nature of oil sludge and its potential environmental effect, its safe disposal poses various difficulties and challenges. In general, no solution can fulfill all the requirements for the reuse and disposal of different types of oil sludge waste. Some treatments may show tremendous promise for fuel recovery or decontamination of non-recoverable remnants. However, their capital or operating costs may be prohibitively costly, or their adoption in a large-scale treatment may be difficult. A wide range of applications and cheap operating costs may be provided by other processing techniques, such as farming and composting, although their microbial breakdown processes may take some time [17].

The properties of the sludge, processing capacity, cost, disposal requirements, and time limitations should all be considered when selecting oily sludge treatment systems. Centrifugation, surfactant recovery, freeze/thaw, froth flotation, and biological sludge treatment are a few procedures that may be better adapted to handle oily sludges with high moisture content. Other processes need pre-treating the sludge to lower its moisture level, including incineration, pyrolysis, and stabilization/solidification. Fig. 2 compares oily sludge treatment and its application level and the average oil recovery rate percentage extracted from different papers [50,57-63]. Because technology selection incorporates several factors, assessing the overall performance of available technologies is challenging. Some multi-criteria assessment analysis methodologies can aid in developing a standard technology evaluation system and selecting the most appropriate disposal methods by researchers [17].

The advantages of many new oily sludge recovery and treatment systems include speed, high efficiency, and energy savings. At the same time, the disadvantages involve high maintenance and operating cost, high running cost, low efficiency, and environmental pollution [50]. Researchers' efforts and accomplishments have been effective in improving efficiency and lowering processing costs. However, most novel approaches are still in the laboratory, far from pilot testing or large-scale technical implementation. Researchers will thus focus on a long experiment to explore new technologies [64].

■ APPLICATIONS OF OIL SLUDGE

The utilization of oil sludge as a secondary raw material appears to be one of the primary developments in the processing of oil sludge [65-68]. This strategy improves the environmental condition in oil refining zones leads to the most efficient use of natural resources.

The selection of the most appropriate technique for the disposal of oil sludge is a difficult task. The disposal and neutralization of highly hazardous waste generated in large cities present several challenges to improving environmental safety. Second, the adaptation and selection of technology for a particular location or territory are determined by the morphological and quantitative composition of the generated waste.

In general, the appropriateness of oil sludge for secondary raw material should be established by its composition, characteristics, and environmental danger. Oil sludge is commonly processed in batches according to

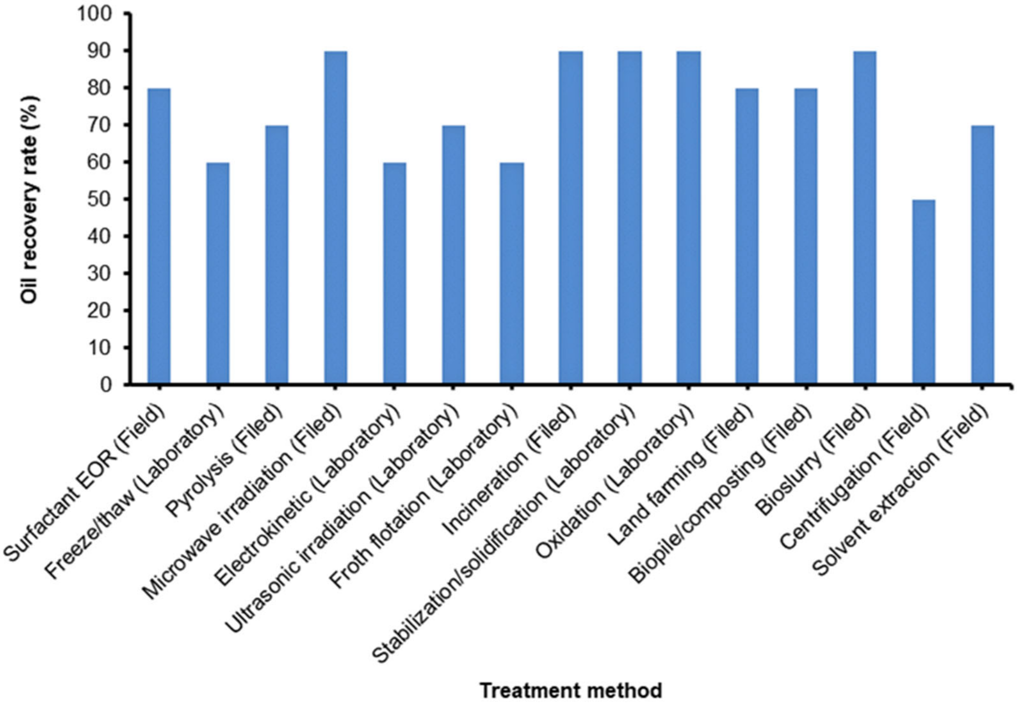

Fig 2. Comparison of oil sludge treatment and disposal methods

to the method used to create it. This approach to the existing situation solves the environmental and rational usage of oil sludge problems. Whether or not oil sludge causes pollution depends on some factors, many of which have to do with its composition and physical characteristics. The processing technique is determined by the consistency of the oil sludge and the organic matter, mechanical impurities, and water content [69-71].

Oil sludge applications are classified based on their technological nature and composition. For example, in the application of road construction, the types of oily waste used are oil sludge refinery (organic part 8–10%; mineral part 70–75%, and water 17–20%) and oil production sludge (organic part 6–40%; mineral part 50–87% and water 5–10%). Another application is oil sludge condensed (organic part 20–25%; mineral part 55–65%; and water 10–25%) and oil sludge refineries (organic part 13–28%; mineral part 59–77%; and water 10–22%) are the types of oily waste used in the application of construction materials. Additionally, the types of oily waste used in the fuel industry are refinery liquid waste (organic part 60–90%; mineral part 5–10% and water 10–20%) and waste from the production of petroleum oils (organic part 77–90%; mineral part 10–14% and water 4–7%). Moreover, the types of oily waste used in bitumen production are the top layer of the acid tar storage (organic part 9–15%; mineral part 65–78% and water 11–26%) and bottom acid tar (organic part 20–26%; mineral part 54–69% and water 18–20%) [69].

CONCLUSION

The three essential components of oil sludge are, as previously stated, the organic portion, the mineral portion, and water. Depending on the type of oil, the organic sludge-mass fraction ranges from 6 to 90%; the mineral sludge-mass fraction ranges from 5 to 87%, and water content ranges from 4 to 25%. Oil sludge with a high mineral concentration is used in road construction, building material manufacturing, and bitumen production. Oil sludge with a high organic matter concentration is used as a fuel component in the petroleum industry.

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AUTHOR CONTRIBUTIONS

All authors played significant roles in the conceptualization and design, data gathering, and analysis. They also contributed to the article's development and revision and decided to submit it to the current journal. They approved the final version to be released and agreed to be responsible for all parts of the effort.

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