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REVIEW

Thermochemical Conversion of Oily Sludge, Composition, Hazards, and Treatment Strategies: An Overview

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ABSTRACT

Oily sludge (OS) is considered one of the most complex by-products produced in the petroleum industry as an unwanted. This work aims to provide a comprehensive review focused on the dangers of stockpiled OS, its sources and contents, as well as its complex composition. This study also analyzes the global annual generation and stockpiling quantities of OS. Therefore, the thermochemical conversion, hydrothermal carbonization (HTC) char produced via HTC called hydrochar, pyrolysis (PY) char produced via PY named pyro-char, and microwave (MW) processes, while the char produced MW labeled micro-char. In conclusion, this study will facilitate researchers and provide a clear understanding of OS making it easy to recovered oil, convert, or exploit as fuel, absorbent materials, or fertilizer for soil. Therefore, this research will contribute to preserving the environment and preserving the planet.

Keywords: Oily sludge, Hydrocarbon, Asphaltene, Fuel, Thermochemical conversion

1. Introduction

The "Environment Protection Act and Hazardous Wastes Handling Rules" state that the petroleum sectors produce the largest amount of waste oily sludge (WOS) that damages the environment [1]. Oily sludge is classified as hazardous waste, requiring treatment to render it innocuous [2]. This dangerous sludge typically builds up in oil tanks during the extraction and processing of oil it is a complicated mixture made up of varying amounts of waste oil, water, and mineral elements [3]. The petroleum industry produces an emulsified solid waste called oily sludge WOS that typically consists of water, crude oil, and airborne particles. It poses a serious risk to both human and the environment due to the high concentration of cycloalkanes, the benzene series, polycyclic aromatic hydrocarbons (PAHs), and other toxic and dangerous compounds. As a result, it must be treated to lessen its toxicity, with ongoing expansion of the oil sector, as shown in Fig. 1, the development of unconventional oil and gas fields in the production of oily sludge has increased [4]. The harmlessness, resource recovery, and reduction treatment of oily sludge, however, have now emerged as one of the major issues in the petroleum sector as a result of the consistent rise in the consumption of petroleum products in recent years, as shown in Fig. 2 [4]. Over the past ten years, researchers have creatively created a variety of disposal and treatment methods in response to stricter environmental legislation and rising public concern about the environmental harm caused by traditional methods of (OS) disposal [5]. As a study mentioned in Figs. 1 and 2 the oil production has been increased up to 8000 million tonnes if 2020 [4], in same content [5] studies demonstrate the daily consumption of oil increased 200,000 parallels per day in same year. These pointers its concern which lade to the researchers to deeply.

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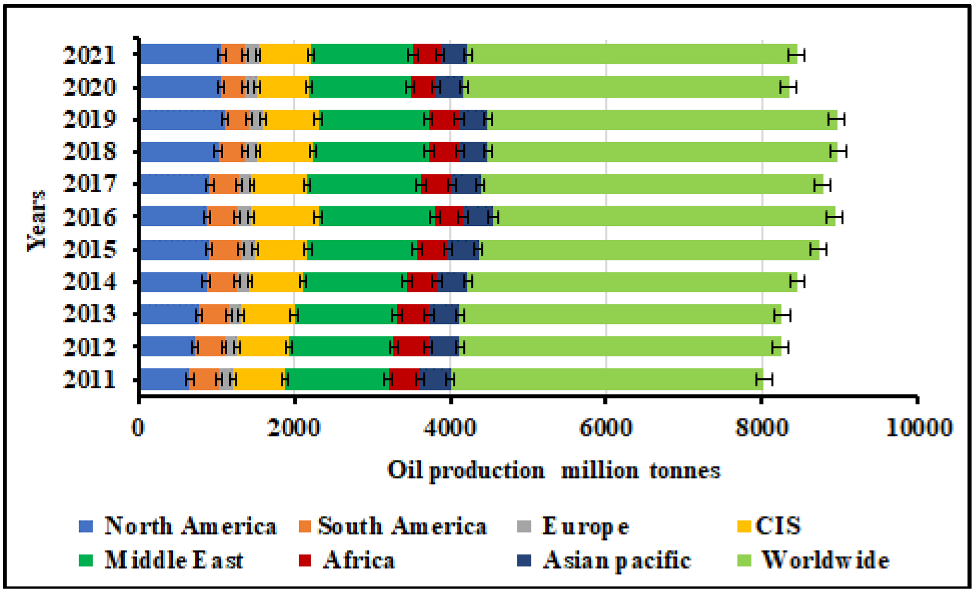

Fig. 1. Oil production in recent years worldwide [4].

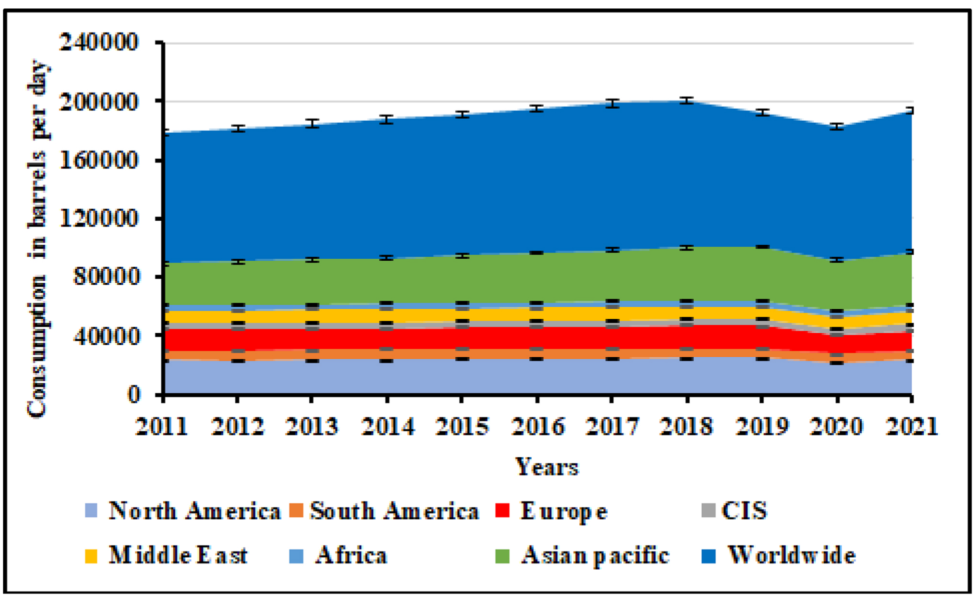

Fig. 2. Daily consumption of oil in recent years worldwide [4].

The challenges faced this work the complex composition of oily sludge which make it difficult to convert into value added materials or cannot forecasting negative results. [6] studies demonstrate and called the hydrothermal carbonization process (HTC), pyrolysis process (PY), while microwave process was called (MW).

This review provides a comprehensive overview of oily sludge (OS) treatment methods, focusing on thermochemical conversion processes such as hydrothermal carbonization (HTC), pyrolysis (PY), and microwave (MW) techniques. The novelty is somewhat limited due to the review nature of the work. The manuscript synthesizes existing knowledge rather than presenting ground breaking findings. However, the comparative analysis of HTC, PY, and MW processes for OS treatment is a strength, as it consolidates scattered research into a unified discussion [7]. Researchers can highlight gaps in current research more explicitly and this work a novel framework or hybrid method combining HTC, PY, and MW for future research. in this work thorough coverage of OS composition, hazards, and thermochemical treatments. Useful tables summarizing data (e.g., metal concentrations, char yields). Clear relevance to environmental and industrial applications. In same content regarding the chars produced during these processes were called (char produced via HTC is hydrochar, her char produced via PY is pyro-char, while the char produced MW process named micro-char). The aims of this review paper were to provide a comprehensive review focused on the dangers of stockpiled OS its sources and contents, as well as its complex composition, to characterize raw oily sludge, using the physicochemical properties such as, Brunauer-Emmett-Teller (BET), thermogravimetric analysis (TGA). Fourier transform infrared (FTIR). The energy characteristics like high heating value (HHV), proximate, element composition, and atomic ratios.to determine its suitability for conversion into solid carbon fuel via different thermochemical conversion processes like; HTC, PY, and MW processes.

2. Waste oily sludge

Waste oily sludge is made up of a complicated mixture of suspended solids, water-in-oil emulsions, and oil-in-water emulsions [8]. It is a hazardous substance that has an adverse effect on both human and the environment [9]. WOS is a type of common and useless solid waste that is mostly created during the extraction, transport, storage, and refining of crude oil [10]. The primary by-product of the petroleum industry is OS which can contain significant amounts of petroleum and heavy metals and, if improperly disposed of, represent substantial risks to both human and the environment [11]. Because of its toxicity and ignitability, OS has been classified by several nations as hazardous waste, the direct disposal of which is severely prohibited. Oil sludge has higher levels of high-quality hydrocarbon compounds than sewage sludge, which means it has less biological activity. In order to reduce sludge volume, heat or chemical material recovery has emerged as one of the most alluring and promising methods for disposing of OS in the future [12].

2.1. Overview of waste oily sludge

Oily sludge typically consists of drilling mud, emulsified solids created during the refining of crude oil, waste oil from wells, and sediments from storage tanks [13]. The recovery of valuable from OS is regarded as a main objective due to its significant economic and environmental advantages [14]. Low oil content sludge is defined as oily sludge with less than 40% recoverable oil. Refinery sludges typically have an oil concentration of > 40% [15]. There are several ways to deal with the OS as mentioned by [16] as following:

1- Hydrothermal carbonization (HTC) for hydrochar production.
2- Thermal hydrolysis (TH) for dewaterability improvement.
3- Hydrothermal liquefaction (HTL) for bio-oil production.
4- Pressurized hot water extraction (PHWE) for hydrocarbon separation.
5- Wet air oxidation (WAO) for biodegradability improvement.
6- Supercritical water upgrading (SCWU) for light oil production.
7- Supercritical water oxidation (SCWO) for complete degradation.
8- Supercritical water gasification (SCWG) for H_2-rich syngas production.

Finding a method of treatment to turn the oily sludge into usable resources is thus necessary. The chemical content and properties of OS differ significantly due to numerous factors such as the form of oilfield, soil composition, and storage conditions; thus, the physicochemical properties of oily sludge from different sources are not identical [17]. The components of crude oil include primarily aliphatic hydrocarbons, aromatic hydrocarbons, asphalt, resins, etc., among which are relatively common benzene, xylene, cycloalkanes, polycyclic aromatic hydrocarbons, and other volatile and refractory organic compounds [18]. Oily sludge belongs to multi-component systems, mainly composed of oil-in-water, or water-in-oil systems, and suspended solids [19]. This is also one of the reasons why oily sludge is highly viscous, poisonous, and acidic, and why its common form is very stable emulsions of oil in water (W / O) or water in oil (O / W) [20].

2.2. Contents of waste oily sludge

When the oil components of the WOS separate, they are collected and reused in the refinery process, and the solid and liquid residues are cleaned before disposal [21]. The WOS typically consists of three components: oil, water, and solids. According to [22], organic phase components of WOS include hydrocarbons, heteroatom compounds, and asphaltene-tar, as well as the water phase and mechanical impurities. These phases typically combine to create a complicated emulsion suspension system that is difficult to separate. Aromatic hydrocarbons, polyaromatic hydrocarbons, and a high total hydrocarbon concentration are among the hazardous components found in OS [23]. Numerous toxic substances are found

Table 1. The main contents in oily sludge [28].

Material	Percentage (%)
Water	55.130
Sediments	9.2490
Asphaltenes	1.9179
Wax	10.5140
Light Hydrocarbons	23.190

in OS, such as polyaromatic hydrocarbons (PHCs), aromatic hydrocarbons, and high level of total hydrocarbons. The four fractions of PHCs and other organic materials in OS include nitrogen compounds that contain sulphur & oxygen, asphaltenes, and aromatics & aliphatic [24]. However, the global composition of oily sludge varies significantly from facility to facility, and from the tank to tank within the same location. Its composition depends on the storage oil composition, storage source conditions, storage times, and storage tank design, and mechanical conditions [25]. [26] studies demonstrate that oily sludge is a rather complex composition; it contains oil in water, oil emulsion water, and solids that are suspended as in Table 1. OS contains poisonous compounds, such as aromatic hydrocarbons, polyaromatic hydrocarbons, and a high proportion of total hydrocarbons. Take for instance, [27] mentioned that main contents of OS are water, sediments, asphaltenes, and wax.

Waste oily sludge is also rich in inorganic contaminants content; among the common inorganic components of OS are zinc (Zn), copper (Cu), nickel (Ni), lead (Pb), chromium (Cr), and mercury (Hg) [29]. [6] mentioned that oil sludge contains the inorganic contaminants as shown in Table 2. Oily sludge is considered a dangerous petroleum compound due to its content of hazardous chemicals. For instance, OS contains a high number of heavy metals such as vanadium (204 ppm), nickel (506 ppm), and Fe (0.6 %), rendering oily sludge hazardous to the environment and to species [25]. That is why it needs to be tackled for the protection of the environment. Table 2 shows the oily sludge for different sample sources.

2.2.1. Asphaltenes

Asphaltenes are mixtures of pentane-insoluble and colloidal compounds such as alicyclic and polyaromatic molecules with alkyl substitutes (methyl groups compounds) [43]. Asphaltenes is the important component of the sludge because it has a high viscosity and can be formed in the form of a mass in the bottom of the oil tanks in the presence of other factors such as plankton and ground roughness and the wall of the tank [44]. Resins and asphaltenes accounts for the stability of WOS emulsion because of their rich hydrophilic functional groups content that can serve as lipophilic emulsifiers and can be treated using solvent extraction method [45].

2.2.2. Nitrogen, sulphur, oxygen compounds

Polar substances such mercaptans, naphthenic acids, pyridines, and thiophenes can be found in the nitrogen, sulphur, and oxygen compounds (NSO) fraction. Less than 3% of the nitrogen (N) is present in OS and the majority of it is found in the distillate residue as part of the asphalt and resin fraction. Sulphur (S) level in OS can range from 0.3 to 10% while oxygen (O) content is often less than 4.8% [48].

2.2.3. Aromatic and aliphatic compounds

Waste oily sludge is composed mainly of oil compounds, mostly the aromatic and aliphatic hydrocarbons. They exist mostly as alkanes, cycloalkanes, xylenes, naphthalene, benzene, toluene, phenols, and numerous polycyclic atomic hydrocarbons PAHs. It is also made up of methylated derivatives of phenanthrene, fluorine, chrysene benzofluorene, anthracene, & pyrene [5].

2.3. Main sources of waste oily sludge

Every stage of the growth of the petrochemical sector involves oily waste. The majority is mostly attributable to the generation of various types of oily sludge such as landing sludge formed during extraction, concentrated sedimentation sludge tank,

Table 2. The concentration of metals in oily sludge.

Zinc Zn (mg/kg)	Lead Pb (mg/kg)	Copper Cu (mg/kg)	Nickle Ni (mg/kg)	Chromium Cr (mg/kg)	Iron Fe (mg/kg)	Mercury Hg (mg/kg)	References
7–80	0.01–0.12	32–120	12–25	27–80	N/A	N/A	[30]
321.7	261.2	105	320	N/A	N/A	31.3	[31]
299	656	500	480	480	200	N/A	[32]
230	38	66.0	42.0	37.2	N/A	N/A	[33]
91	49	55	16	14	5324	N/A	[34]
1299	265	500	175	480	60200	N/A	[32]
7–80	N/A	32–120	17–25	27–80	N/A	N/A	[35]
6100	580	N/A	2700	N/A	N/A	N/A	[36]
110.1	–	28.2	38.5	74.5	–	–	[37]

Table 3. Contents of differente oily sludge samples.

Origin of oily sludge samples	Hydrocarbons %	Water %	Solids %	References
China National Offshore Oil Corporation	30–80	30–50	10–20	[37]
India, Hindustan Petroleum, Mumbai	15–50	30–85	4–46	[38]
Libya, Azzawiya oil refinery	30–50	30–50	10–20	[39]
China, Dagang Oil Field	15–50	30–85	5–46	[40]
China, Wuhan, petrochemical refinery	20–30	30–40	40–50	[41]
China, Heilongjiang, Daqing Field	36.78	13.76	49.94	[42]

Table 4. Components of different oily sludge samples.

Origin of oily sludge	Alkanes Mass (%)	Aromatics Mass (%)	Asphaltenes Mass (%)	Resins Mass (%)	References
China, Shengli Oil Field	26.69	23.53	14.51	35.27	[44]
India, Bhilai, steel plant	40–55	28–33	8–12	10–22.4	[46]
Jordan, Zarka, oil refinery	65.24	14.88	12.18	7.66	[47]

Table 5. Composition variations and standard oily sludge assessment.

Source	Advantage	Evaluation	References
Landing sludge	- Rich in asphaltene, silt, gum, additives, and heavy metals - The high content of water (more than 10%) - The high content of N and P - Low content of H and C - Brown or black appearance	- Larger viscosity - Low calorific value. - High-risk factor	[50]
Tank bottom sludge	- Rich in hydrocarbons, asphaltenes, paraffin, water and inorganic solids, such as sand, iron sulphide, and iron oxide - The high content of C (up to 80%) and H (up to 10%) - The high content of metals	- High calorific value and recovery value - Easy for secondary pollution - Less contact with residents	[51]
Refining sludge	- Rich in benzene series, phenols and additives - High content of bacteria and water	- With irritating odor - Complex properties high viscosity - Low recovery value - Centralized and easy to collect	[52]
Sediment pond sludge	- High moisture content and low hydrocarbon content - High content of N and S.	- High yields - Serious leak - High risk of secondary pollution - Low recovery value	[53]

sludge produced during tank cleaning at the bottom of storage tanks, and oily sludge produced during chemical processing. With joint parameters such sample time, location, and many other characteristics, the oily sludge qualities and composition vary greatly. Its qualities can occasionally change in a wide range, even for the same kind of oily sludge [45]. The oily sludge used in this study is bottom tank oily sludge collected from the North Refineries Company. Table 5 shows the advantage and evaluation of the composition of different types of WOS [49].

2.4. Generation of waste oily sludge

Recent years have seen a huge increase in the amount of waste produced by the petroleum sectors, which is a significant cause of environmental pollution. According to reports, the amount of wastewater produced by the petroleum refining process is roughly 1.6 times the amount of crude oil processed [21]. About two-thirds of the waste produced in petroleum refineries is made up of oily sludge [54]. These wastewaters include dissolved minerals, phenols, and hydrocarbons as shown in Fig. 3.

Benzene, toluene, ethylbenzene, and xylene are examples of common hydrocarbons that are either poisonous or carcinogenic. Produced water, production chemicals, workover waste, and sludge bottom tanks are among the common by-products from production activities [55] as shown in Table 6. The hazardous nature of WOS has been a source of worry due to the danger it can cause to the environment. North refineries company (NRC) Baiji produces around 3000–3500 m^3 of WOS per year, and about

Table 6. Common waste generation in oil industry [55].

Petroleum Waste	Constituents
Produced water	Heavy metals, organic, hydrocarbons, ammonia, H_2S
Produced sand	Heavy metals, hydrocarbons
Oily sludge / bottom waste	Chemicals, hydrocarbons, heavy metals
Contaminated soil	Hydrocarbons, heavy metals, chemicals
Waste lubricants	Heavy metals, and hydrocarbons
Ballast water	Heavy metals, and hydrocarbons
Domestic waste	Organic, solids
Muds, drilling fluids and cutting	Viscosities Biocides, surfactant, metals
Oil spill/leaks	Hydrocarbons, chemicals
Boiler blowdown	Biocides, heavy metals

Table 7. Production of WOS in some oil producing countries.

Countries	Oily sludge sources	Oily sludge quantity	References
Malaysia	Petroleum refinery	3,200 m^3/year	[60]
Iraq	NRC Baiji	3,000–3,500 m^3/year	[25]
Morocco	Petroleum refinery industry	2,000 tons/year	[57]
China	Petrochemical industries	3 million tons/year	[61]
India	Refineries	28,000 tons/year	[35]
Worldwide annually	Various sources	60 million tons	[62]
Worldwide accumulated	Various sources	1 billion tons	[62]

90% of these quantities are disposed of in landfills, which cause an adverse effect on the land in Makhoul Mountains North of NRC [56].

2.5. Quantities of waste oily sludge

All the oil-producing countries of the world have oily sludge production. According to the production theory in force, for every 500 m^3 of processed crude oil, 1 m^3 of oily sludge is produced [57]. In general, one kilogram of crude oil contains 10–20 g of waste residue [58]. Oil production companies produce a huge amount of oily sludge and other unwanted products [59]. Malaysia produced more than 3200 m^3/year of oily sludge from the refinery sector [60]. Moreover, these quantities listed in Table 7 have resulted in the production of more than 60 million tons of oily sludge per year and more than 1 billion tons of oily sludge have been staked worldwide [6].

2.6. General characteristic of oily sludge

2.6.1. Characterization of Malaysia oily sludge

Oily sludge properties are important in petroleum products and must be precisely known in order to know the fuel behaviours and how to deal with it. The study by [63] studies demonstrate the characteristics of the local oily sludge refinery Malaysia (see Table 8). It was found that the sludge contains a high proportion of carbon that can be recovered, as well as deposits and amounts of water. It also shows that oily sludge contains more hydrocarbons materials, which makes oily sludge a useful product and that can be used in energy production after reclamation.

Table 8. Properties of oily sludge refinery [63].

Characteristics	Result %
Moistures content	61.4
Total solid residue (TSR)	35.8
Total organic carbon (TOC)	2.2
Polycyclic aromatic hydrocarbon (PAH)	0.62
Aliphatic hydrocarbon compounds (AHC)	86.26
Diesel range organic (DRO)	84.3
Gasoline range organic (GRO)	0.02
Oil range organic (ORO)	15.68
Other compounds	13.12

2.6.2. Thermochemical properties of oily sludge

Oily sludge obtained from Petronas Melaka in Malaysia was characterized by physical and chemical features. The proximate of oily sludge was found to contain high moisture (78.91%), low ash (5.06%), low volatiles (5.52%), and high fixed carbon (10.51%). The sludge has a calorific value of 23.599 MJ/kg. Despite the high moisture content, the higher heating value (HHV) is high when compared to literature values. The high value of HHV may be associated with the high fixed carbon, low ash content, and the apparent density of oily sludge is 1.08. These results listed in Table 9, the study of the characteristics of oily sludge extracted from Petronas showed the necessity of separation of heavy metals in it, and the gasification of lighter ones as exhaust gas on the other before dealing with this oily sludge [23].

Table 9. Charactrization of oily sludge [23].

Property	Results
Density (g/cm^3)	1.08
Moisture content (%)	78.91
Volatile matter (%)	5.52
Ash (%)	5.06
Fixed carbon (%)	10.51
Calorific value (MJ/kg)	23.599

2.6.3. Proximate and ultimate analysis of Malaysia oily sludge

Characterization of wet and dried oily sludge from a petroleum refinery in Melaka [64]. The proximate analysis, moisture content of sludge is very high (75.3 wt%) which would decrease the heating value. However, the higher heating value (HHV) of 20.5 MJ/kg of sludge in this study is higher than HHV in other petroleum sludge probably due to the hydrocarbon content in the oily sludge. It is also found that volatile matter is high at 58.6 wt % where it can be converted into valuable products via material decomposition. Relatively high content of oxygen element (39.7 wt %) contributed to the low value of HHV and possible highly oxygenated liquid product during pyrolysis. Metal content analysis revealed high content of Fe (1,850 ppm) and also significant amount of Al, Mg, Zn and Na which categorize the waste as schedule waste. Hence, good penetration can be achieved by microwave energy during microwave thermal treatment process to allow material decomposition but the energy conversion will be occurred fairly. Table 10 shown proximate analysis of oily sludge sample. Table 11 shown impossible find ultimate analysis of oily sludge sample.

Table 10. Proximate analysis and HHV of oily sludge [64].

	Proximate analysis				
Oily sludge	M %	VM %	Ash %	FC %	HHV MJ/kg
Wet sample	75.3	9.9	6.7	–	–
Dried sample	3.1	11.4	27.3	59.9	20.5

Table 11. Ultimate analysis of oily sludge [64].

	Ultimate analysis				
Oily sludge	C %	H %	N %	O %	S %
Wet sample	–	–	–	–	–
Dried sample	59.9	4.0	1.2	3.3	3.3

2.6.4. Calorific value of oily sludge

The thermodynamic study of the results of the combustion process attempts to assess the quantity of heat transmitted to the atmosphere by fuels [65]. In chemical and fuel characterization, higher heating value (HHV) is very important. In this analysis, however, the higher heating value (HHV) of 20.5 MJ / kg of oily sludge can be due to the hydrocarbon content in the oily sludge [66]. Studies demonstrate [67] that the HHV of OS is 23.599 MJ/kg. Although the moisture content is high, when compared with literature values, the HHV is high. The high HHV may be correlated with low ash content and high fixed carbon.

2.6.5. Proximate and ultimate analysis of oily sludge

The quantitative examination of tests such as percentages of moisture, volatile matter, ash, and fixed carbon is included in the proximate analysis of fuel [68]. A higher moisture content decreases calorific value, raises transportation costs, and causes a significant amount of heat to be lost through evaporation. High levels of volatile matter result in lengthy, smoky flames and a reduction in calorific value. The calorific value of the fuel is reduced as the percentage of ash increases. High carbon content will produce hydrochar of high quality and improve calorific value, as shown in Table 13. The ultimate analysis of oily sludge show that it is high in the content of carbon (85%), low in inorganic content, and relatively high in oxygen content (up 35.52%) which can promote their interaction during co-carbonization treatment [69]. Table 12 shows the proximate analysis of oily sludge. Table 13 shows the ultimate analysis of oily sludge.

2.6.6. Thermal stability analysis of oily sludge

The thermogravimetric analysis reflects the rate of mass loss versus temperature as seen in Fig. 3. The temperature range between 200 and 500°C exhibits the greatest mass loss. At this time, the mass-loss rate curve shows two different peaks at 390 and 450°C. Due to the volatilization of heavy oil (C20+) and diesel oil (C12 to C20), the mass-loss rate curve at this point exhibits two different peaks at 390 and 450°C [75]. The OS sample exhibits a further weight loss of 0.6 mg, or 10% of its initial mass, in the temperature range of 500 to 1200°C. This could be due to heavy oil component carbonization as well as mineral decomposition. The rise in weight loss at 600−800°C may be attributed to catalytic tar cracking, reforming the release of more gaseous materials and a potential reduction in iron oxides gas [76].

2.6.7. FTIR spectrum of oily sludge

Fig. 4 compares the FTIR spectra of the oily sludge with other samples. The distribution of functional groups in the two types of pyrolysis oily sludge is comparable. FTIR spectra are used to confirm the presence of aromatic compounds in the pyrolysis oil. Fig. 4 shows the FTIR spectra of various oily sludge samples, including (a) asphaltene, (b) clinoptilolite, and (c) cleansed solids. By comparing curves,

Table 12. Proximate analysis of different source oily sludge.

	Proximate Analysis				
Oily Sludge Samples	M %	V %	Ash %	FC %	Reference
Malaysia, Melaka, refinery plant	75.3	8.10	28.2	60.6	[70]
China, Nahai, solid waste disposal Co.	33.4	4.80	50.1	11.7	[71]
China, Nahai, solid waste disposal Co.	1.5	89.6	0.2	8.7	[72]
China, Shaanxi province, oil field	10.26	32.72	54.6	2.43	[8]
China, Shengli, oil field	1.62	21.33	71.7	5.37	[8]
China, Liaohe, oil field	34.84	48.59	7.79	6.78	[8]
China, Tianjin, national offshore oil	36.06	26.10	33.08	4.76	[37]
China, Tahe, oilfield	10.40	72.12	15.62	1.86	[37]
China, Yumen, oilfield	34.17	30.44	32.60	2.79	[37]
India, Tami lnadu, petroleum refinery	17.50	60.42	16.62	5.46	[73]
Chennai, Manali, oilfield	9.8	94.8	1.3	5.2	[74]
Chennai, Manali, oilfield	4.4	98.1	19.6	1.9	[74]

Table 13. Ultimate analysis of different source oily sludge.

	Ultimate analysis					
Oily sludge samples	C %	H %	N %	O %	S %	Reference
Malaysia, Melaka, refinery plant	45.0	6.6	7.0	39.7	1.7	[70]
China, Nahai, solid waste disposal Co.	79.4	7.6	0.5	6.2	6.3	[71]
China, Nahai, solid waste disposal Co.	85.5	9.8	0.3	0.4	2.3	[72]
China, Shaanxi province, oil field	23.68	3.07	1.16	12.25	3.16	[8]
China, Shengli, oil field	18.8	2.69	0.28	6.05	0.65	[8]
China, Liaohe, oil field	42.45	8.07	1.21	30.34	1.42	[8]
China, Tianjin, national offshore oil	22.65	3.44	15.60	15.60	1.28	[37]
China, Tahe, oilfield	7.34	1.23	18.54	18.54	0.55	[37]
China, Yumen, oilfield	20.56	3.11	0.40	31.11	0.54	[37]
India, Tami lnadu, petroleum refinery	49.75	5.51	0.36	38.98	5.4	[73]
Chennai, Manali, oilfield	73.8	11.3	0.5	8.6	0.5	[74]
Chennai, Manali, oilfield	39.4	5.1	8.4	1.7	6.3	[74]

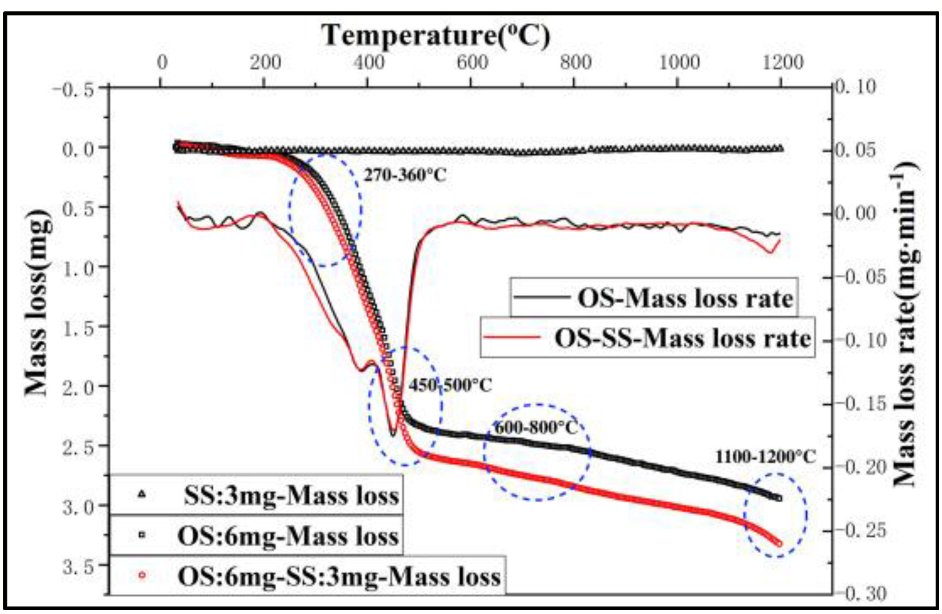

Fig. 3. TGA and DTGA curves of oily sludge [76].

a and c, we can determine that there were crude oil components on the solid surface, which is supported by the bands that are distinctive to asphaltene [77]. At 2923 cm^{-1} and 2853 cm^{-1}, the (-CH2-) and (-CH3) stretching vibrations of the alkyl chain are very well established. In cyclic ketones, ester, aldehydes, or carboxyl, the (C=O) stretching vibrations at 1700 cm^{-1} represent the carboxyl functional group.

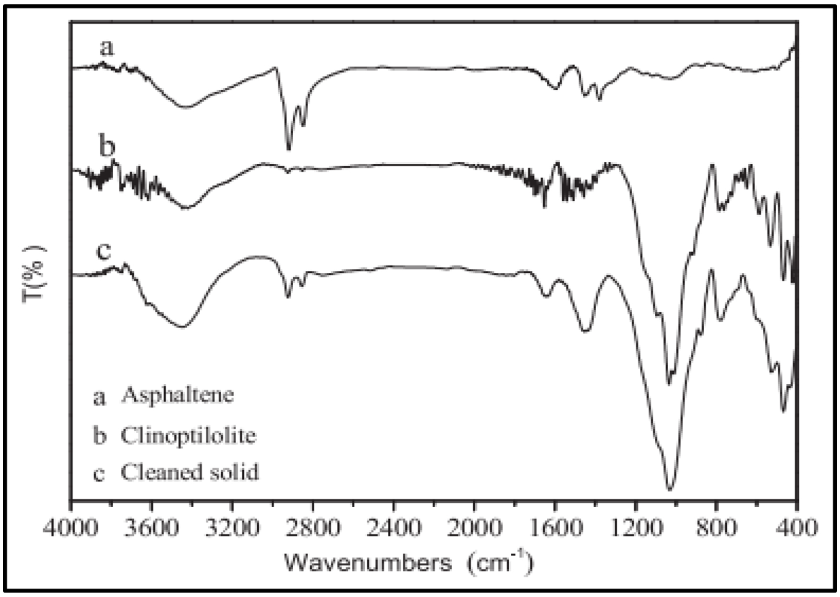

Fig. 4. FTIR Spectra of oily sludge [78].

Table 14. Textural properties of OS, hydrochr, pyrochar, microchar [6].

Sample name	V m cm^3/g	Total pore volume cm^3/g	BET surface area m^2/g	Pore volume m^2/g	Average pore volume (nm)
OS	0.00413	0.05626	29.56	41.71	7.43
Hydrochar	0.00236	0.05744	31.77	32.88	10.10
Pyro-char	0.02397	0.02963	81.33	93.65	1.73
Micro-char	0.06940	0.09535	238.52	283.65	1.88

Additionally, the aromatic rings' (C=C) stretching vibrations form in superposition with the clinoptilolite bands at 1450 cm^{-1} and 1641 cm^{-1}, just as the (C-O) stretching vibration at 1100 cm^{-1} and the (-OH) and (-NH2) stretching vibration at 3444 cm^{-1} [78]. These findings showed that some heavy crude oil constituents, such as long-chain alkanes and PAHs in asphaltene were still getting adsorbed on the cleaned solid surfaces. Numerous research findings also demonstrate that the adsorption of asphaltenes on the surface of various minerals is a complex multilayer adsorption process.

2.6.8. BET of oily sludge and chars

Brunauer-Emmett-Teller (BET), The textural properties (e.g., BET surface area, Langmuir surface area, mean pore diameter, and total pore volume). [6] studies demonstrate as in Table 14 displays the textural characteristics of the raw OS and the produced chars at the optimum condition (ASTM B922) [79]. According to Table 14, micro-char had a BET surface area of 238.52 m^2/g, which was significantly higher than OS (29.56 m^2/g), hydrochar (31.77 m^2/g), and pyro-char (81.33 m^2/g). The increase in BET surface area for micro-char was 8 times more than that of OS, 8 times greater than that of hydrochar, and 2 times greater than that of micro-char. This discovery relates to the MW heating process, which, as a result of the MW's uniform heat distribution, controllability, and high heating accuracy, significantly affects the BET surface area [80]. Additionally, OS, hydrochar, pyro-char, and micro-char have mean pore diameters of 7.43 nm, 10.1 nm, 1.73 nm, and 1.88 nm, respectively. According to IUPAC categorization, these results showed that the created chars have a microporous structure (mean pore diameter < 2 nm) [81].

3. Potential hazard of waste oily sludge

The PAHs, heavy metals, phenols, anthracene, pyrene, and other poisonous substances in oily sludge are what pose the greatest threat to the environment and human health. If left untreated for a long period of time, prolonged stacking of oily sludge can seriously contaminate the soil, groundwater, and environment [19]. The thermochemical methods of OS reduce them quantitatively (e.g., PAH destruction efficiency).

The treatment of oil sludge has become a global concern and has garnered a lot of attention from relevant scholars due to the composition of oily sludge and its severe harm to the environment [82]. In an effort to solve this issue, several researchers have tried to transform oily sludge into valuable goods, with the production of activated carbon being the most frequently researched of them. However, the reported activated carbon made from oily sludge is still unsuitable for industrial applications, and there are certain environmental dangers [83]. In this study, it was found that converting oily sludge to activated carbon is not a profitable process. Therefore, the researcher resorted to converting sludge to solid carbon fuel hydrochar due to the high carbon content and the basis for the formation of oily sludge is fuel oil. However, due to the complex composition of oily sludge and the hazard it poses to the environment, Ammar [84] mentioned that oily sludge affects the environment in the following aspects:

1- Contamination of the soil: The disposal of oily sludge and its hazardous components significantly contaminates the surface, and therefore the groundwater.
2- Occupies vast areas of land: Since there is not yet a completely effective process for reclaiming oily sludge, it builds up in an environment and consumes more and more land, in addition to methane gas; this would create huge quantities of toxic gas that may cause fire and explosions.
3- Pollution of water bodies: Oily sludge of natural precipitation flows into the rivers, streams and the smaller particles may be carried by wind and deposited in surface water.
4- Toxic components: Chemical contaminants, heavy metals, and other hazardous chemicals, long-term storage of oily sludge in oil tanks, raises the risk of runoffs due to contamination or other forms of incidents that could cause major releases to the atmosphere.
5- Atmospheric pollution: Being biodegradable, exposed oily sludge to appropriate temperature and humidity could be degraded by microorganisms easily, releasing many harmful gasses.

4. Oily sludge treatment

Oily sludge disposal in refinery operations is the first step in the reclamation process. Even after being de-oiled, oily sludges cannot be completely remedied and must instead be disposed of in landfills. There are many approaches to treating oily sludge, including two basic ones: disposal and oil recovery with specific sub-approaches. All techniques for dealing with oily sludge, as seen in Fig. 5 [5], promise to improve it. Finally, based on this research, we should discover a new technical solution that is quick, affordable, and safe for dealing with oily sludge so that it can be transformed from a dangerous substance into a valuable product [85].

4.1. Disposal of oily sludge

Petroleum sludge is treated using disposal techniques after all usable oil and hydrocarbons have been recovered. Incineration, oxidation, solidification/stabilization, and biodegradation are a few of the techniques used. The negative impact of this hazardous waste on the environment has been lessened by the deployment of these treatment technologies to a greater extent. Additionally, a variety of traditional and contemporary procedures has been used to remediate the oily sludge from petroleum refineries. [86] described using an incinerator to treat oily sludge that was unsuitable due to solid hydrocarbon-based components.

4.2. Oil recovery from oily sludge

The petroleum industry is the recovery of the valuable oily phase back to the refining scheme and disposal methods of the remaining fraction without any valorization. To recover the oily phase, solvent extraction, and centrifugation have been used. Nevertheless, novel approaches to introduce more commercially achievable options and products to reduce socio-economic and environmental problems associated with its current treatment must be studied. Oily sludge may be processed to produce useful products or as a feedstock for energy generation. Different valorization technologies, such as recovering the oily phase or the production of a char or activated carbon can be found in the literature. However, all these treatments are only focused on one of the phases of the sludge (oily or solid), which means that only (15–50 %) of the oily sludge is valorized, and the remnant (50–85%) is disposed of without taking advantage of the potential resources contained in it [87].

Water, mud, emulsified solids produced during the refining of crude oil, and sediment in the storage tank bottom are all components of oily sludge, which is mostly produced during the production, refining, storage, and transportation of petroleum. The oil recovery treatment from oily sludge has recovered a high percentage of oil up to (50%) and minimized the risk of solids to about (30%) using the method of energy recovery. Recovery is one of the alternative approaches to dealing with oil sludge. The process of recovery will lessen the amount of sludge produced by the oil industry, lessen environmental damage, and preserve the economy's supply of energy resources [86].

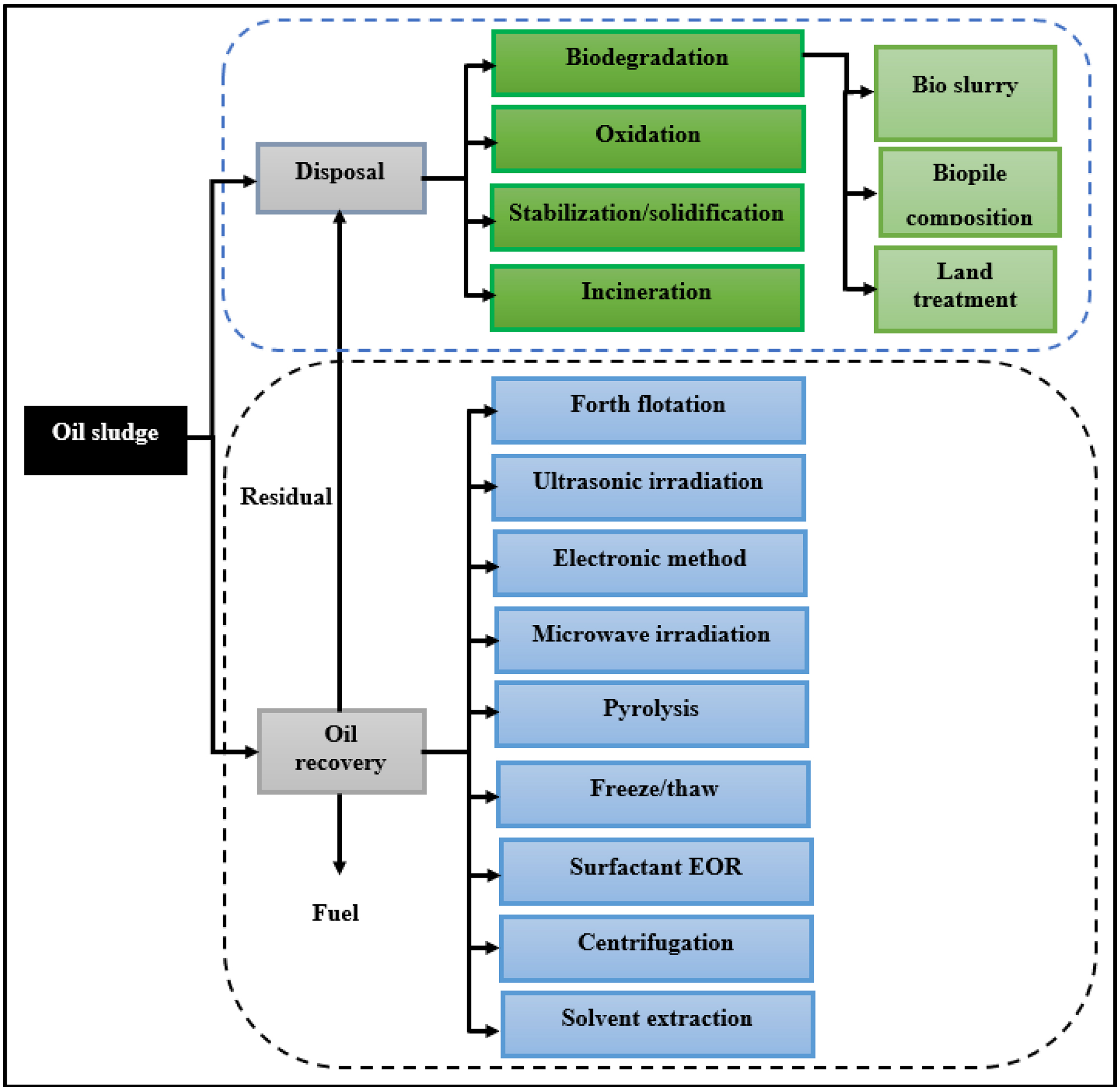

Fig. 5. Current oily sludge treatment methods [5].

5. Hydrothermal processes

In order to create energy materials and boost energy density, hydrothermal treatment includes heating aqueous slurry of raw materials and organic waste under pressure. High moisture and ash content raw materials can be treated hydrothermally without the necessity for pre-drying [88]. The HTC process is one of the thermochemical conversion methods that has caught the attention of researchers since it can directly handle wet raw materials. As shown in Fig. 6, waste is often transformed into valuable materials such carbon in the form of hydrochar, as well as non-condensable gases (mainly CO_2) and water phase products [89]. HTC process transforms raw material to value-added products at mid-pressure in a humid environment [90, 91]. HTC process has regained significant interest in recent times due to the need for efficient pre-treatment technologies and the advantages of HTC over other thermochemical pre-treatment methods, such as the conversion of wet feedstock to hydrochar [92]. The hydrothermal process is in different forms based on the operating conditions (temperature, and pressure), the product state yield solid liquid or gaseous, hydrothermal carbonization (HTC) from 150 to 250°C, pressure 0.5 to 4 MPa [93]; hydrothermal liquefaction (HTL) 150 to 350°C, pressure 3 to 6 MPa, [94, 95]; hydrothermal gasification (HTG) above 350°C, and higher pressures beyond 8 MPa, [96].

5.1. Hydrothermal carbonization process

A closed system, high autogenous pressure between 2 and 10 MPa, wet material as the feedstock, and

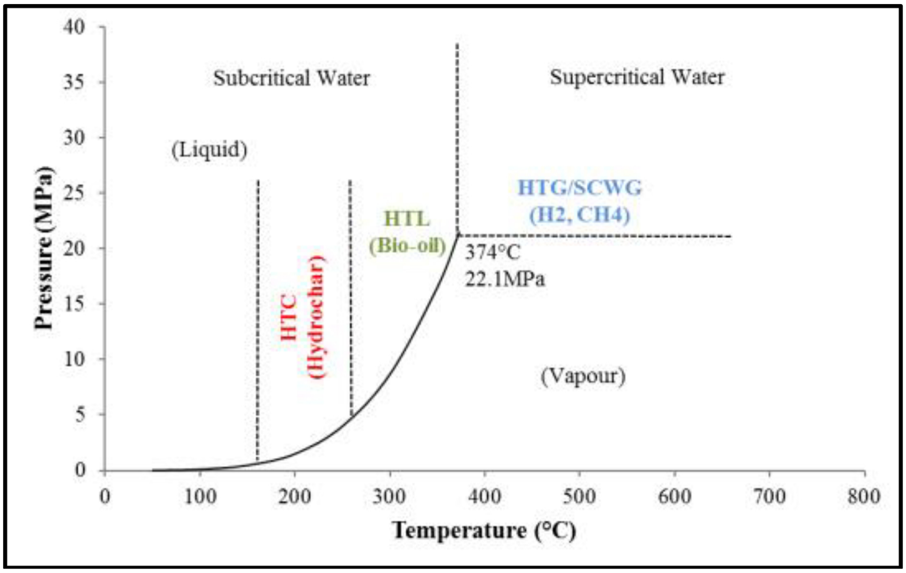

Fig. 6. Temperature and pressure of hydrothermal methods [97].

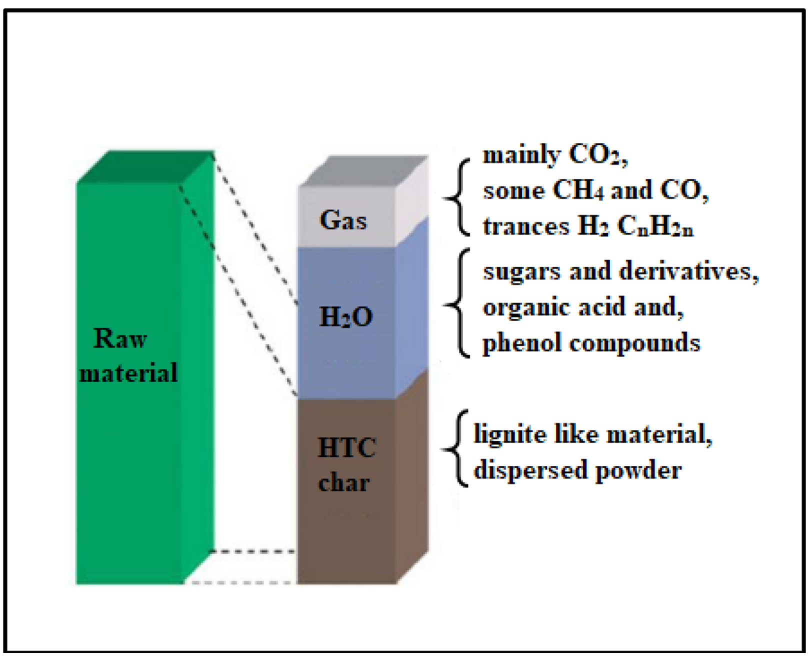

Fig. 7. HTC products by range of temperature [105].

a temperature range between 150 and 280°C are all required for hydrothermal carbonization (HTC) [98]. Wet torrefaction, also known as HTC, is a thermochemical process that converts organic feedstock into a solid product with a high carbon content. The method involves mixing water into the raw material and heating it in a closed system at temperatures between 180 and 260°C with reaction periods between 5 minutes and 4 hours under pressures between 2 and 6 MPa [99]. The reaction pressure, which corresponds to the reaction temperature with the saturation vapor pressure of water (subcritical-water), is often not regulated in the process and is autogenic [97]. The HTC technique has drawn a lot of attention because it uses water, which is non-toxic, affordable, environmentally friendly, and also present in green biomass [91].

In the HTC process, the breakdown of the feedstock results from serial simultaneous reactions, including dehydration, hydrolysis, dramatization, decarboxylation, and re-condensation [100]. HTC can be defined as two reactions (dehydration and decarboxylation) of feed material to raise carbon content and get a higher calorific value [98]. As in Fig. 7, HTC

Table 15. HTC processes summarized of oily sludge for hydrochar production.

Oily Sludge Samples	Temperature °C	Hydrochar Yield %	HHV MJ/kg	References
China, Nanyang, oil field	240	79.5	21.71	[106]
India, Chennai, petroleum refinery	250	65.3	–	[107]
India, Chennai, petroleum refinery	250	51.73	26.79	[73]

main product is a solid called hydrochar which also creates liquid (aqueous soluble) and gas (mainly CO_2) by-products depending on the reaction temperature range. According to study [101], the HTC process's conversion procedure creates hydrochar, a carbon-like fuel with well-known qualities that can be handled easily from waste leftovers thanks to its high moisture content [98]. The HTC process is highly effective for converting wet raw material into hydrochar because it does not require prior drying of the materials, thus saves energy that would have been needed to dry the raw material when the solid, liquid and gaseous products were cooled, filtered, phase-separated and distilled from the process [102]. HTC processes a range of feedstock, including herbaceous, plastics material, and woody feedstock [103]. Table 15 shown the summarized past results of hydrothermal carbonization processes of oily sludge for hydrochar production and effect of reaction temperature on the hydrochar yield and HHV recovered. [104] studies demonstrate the best residence time was 120 min.

5.1.1. Advantage of HTC process

The HTC process of treating feedstock has a number of benefits over other thermochemical treatment processes (torrefaction or combustion). In contrast to other thermochemical processes, which might take days or weeks, this reaction only lasts a few minutes to a few hours. The output products are sterile and hygienic as a result of the high process temperatures that also destroy microorganisms and inactivate other potential pollutants like medications [91]. Additionally, HTC is regarded as a successful method for reducing greenhouse gas emissions. HTC is actually the most effective method of converting carbohydrates into products with higher carbon contents or other burnable fuels. Some of the carbon that was originally present in the feedstock is changed into CO_2 and lost to the atmosphere when it is composted, anaerobically digested, or fermented. With HTC, however, the majority of the carbon that was initially present in the substrate remains bonded to the finished product [108]. Additionally, [109] studies demonstrate the benefits of biomass in the HTC. In general, hydrothermal carbonization occurs at temperatures that are substantially lower than those of pyrolysis, gasification, and flash carbonization. The fact that water is the solvent eliminates the need for pre-drying. Some pollutants, including CO_2, sulphide oxides, and nitrogen oxides, dissolve in water, producing the corresponding acids and/or salts and possibly eliminating the need for additional treatment of air pollution.

5.1.2. HTC reaction mechanism

The HTC process, which is exothermic, reduces the feed's oxygen and hydrogen concentration in accordance with the molecular (O/C and H/C ratio) ratio [110]. HTC is the process of combining dehydration and decarboxylation to increase a material's carbon content and increase its calorific value [111]. As the residence time and reaction temperature were raised, the hydrochar's carbon, hydrogen, and nitrogen (CHN) contents rose [112]. The hydrochar's hydrogen and oxygen contents decreased while its carbon content rose [113]. These reactions are in line with how feedstock behaved during the HTC process, which has been attributed to a variety of processes including dehydration, deoxygenation, and decarboxylation [114]. After the HTC process, the H/C and O/C ratios were decreases mainly due to the decarboxylation process [115]. The study by [98] studies demonstrate the mechanism of HTC reaction to include decarboxylation, polymerization, aromatization, dehydration, condensation and hydrolysis. Furthermore, in HTC processes, the main reactions are decarboxylation and dehydration [116].

5.2. Pyrolysis process of oily sludge

Pyrolysis is classified into two types based on heating methods - conventional pyrolysis (CP) and microwave pyrolysis (MWP) [117]. A thick carbonization layer that forms on the sludge's surface during convectional pyrolysis hinders heat transport, causing the sludge to have a high surface temperature and a low core temperature [118]. Traditional and cutting-edge treatment techniques such as solvent extraction, combustion, and pyrolysis were frequently used to treat the oily sludge, with an emphasis on either resource recovery or environmental effects [10]. Particularly, the performance balance of pyrolysis technology in terms of resource recovery and

Table 16. Pyrolysis processes summarize of oily sludge for pyro-char productio.

Oily sludge samples	Temperature °C	Pyro-char Yield %	HHV MJ/kg	References
China, Wuhan, iron and steel Co.	600	57.48	34.59	[123]
China, Daqing, Province, Oilfield	700	58.73	–	[124]
Iraq, NRC Baiji	700	57.30	17.14	[125]

environmental effects has raised questions [119]. Pyrolysis has been deemed the most suitable approach for the treatment of petroleum oily sludge based on economic, social, and treatment difficulties considerations [120].

Pyrolysis is an effective method for handling oily sludge in a safe manner with minimal environmental effects, but pollutant emissions during the pyrolysis process remain a significant challenge [121]. Pyrolysis is a thermochemical treatment method that can turn oily sludge into products (e.g., condensable liquid oil, non-condensable gas, and solid residue)" with additional value when used in anaerobic environments [122]. There is different pyrolysis based on conditions such as slow, fast, and flash pyrolysis as detailed in the next section. Table 16 shown the summarized of pyrolysis processes of oily sludge for pyro-char and effect of reaction temperature on the pyro-char yield and HHV recovered.

5.2.1. The slow, fast, and flash pyrolysis processes

Pyrolysis can be classified as slow, rapid, or flash depending on the conditions. Each pyrolytic procedure has advantages and disadvantages. The temperature and residence time conditions utilized in pyrolysis affect the composition and yield of char [126]. As a result, understanding the pyrolytic processing conditions is critical for determining the appropriate pyrolytic method for getting the desired char quality and production. Temperature, residence duration, pressure, raw material nature, and other reactor variables all influence the primary products oil, char, and gas. Furthermore, by selecting the optimal pyrolysis method and feed materials, the concentration of solid, liquid, and gas can be adjusted [127]. In general, slow, fast, and flash pyrolysis depends on the heating rate, feed amount, residence time, and temperature.

5.2.2. Slow pyrolysis processes

In slow pyrolysis, the temperature is increased slowly with a slow heating rate. Vapours that persist in the slow pyrolysis reactor between 10 and 60 min are majorly used for char production. The hearting rate in the slow pyrolysis reactor is in the range of 0.1 and 1°C / second and char, oil or gas can be produced as the main product based on the process conditions [128]. During the process, the raw materials placed in the reactor and heated using the electric furnace at a rate of < 10°C / second. The slow pyrolysis process is carried out in the absence of O_2 gas. The N_2 gas is used as the sweeping gas to avoid oxygen presence inside the reactor. Slow pyrolysis is greatly beneficial for biochar yield as other liquid and gaseous products are obtained at a minimal level relative to char [129]. The raw material is degraded at successive stages of slow pyrolysis at temperatures ranging from 400 C to 500 C. At first, bond breakage and water content are removed. The creation of pyrolysis decomposes lipids and carbohydrates in the second stage. Carbon-rich residues are generated at the end. The yield of organic liquid was between 50 and 70 % w / w when the temperature range was between 450 and 550°C [130].

5.2.3. Fast pyrolysis processes

Fast pyrolysis occurs under reaction conditions of moderate temperature, moderate rate of heating, and short residence time, with oil (liquid fuel) as the primary product [130]. Fast pyrolysis is a technique for thermally breaking down diverse biomass types in the absence of air or oxygen to produce three sorts of products based on their nature, namely solid, liquid, and gas. This technique produces pyrolytic gas. After cooling and condensing, a dark brown homogeneous liquid with a high heating value known as oil is formed. The fast pyrolysis of raw material yields three primary products (char, oil, and gas). Oil can be used as a fuel in engines and boilers, and it can also be utilized to generate electricity and heat through combined heat and power plants. This process's temperature range is normally 350–600°C, however the temperature for maximum yield is most commonly around 500°C. The residence time is reduced to about 2 seconds, while the heating rate is increased [131].

5.2.4. Flash pyrolysis processes

The principal result of flash pyrolysis (or extremely rapid pyrolysis) is oil, which is produced at a high rate of heating with a very tiny feed particle size of less than (2 mm) and a very short residence period [132]. In general, pyrolysis produces three basic products: oil, gas, and char. The ultimate targeted products determine which of these three methods of pyrolysis is used. That is, the pyrolysis process can be optimized to produce char (coal), oil, or gas. Several researchers have used slow pyrolysis to produce bioenergy (oil, char, and gas) from raw materials [133].

5.2.5. The influence of operating parameters

Among the products of OS pyrolysis are sludge, oil, and residue. Considering that the two main goals are oil recovery and disposal of non-hazardous waste, oil yield and oil content in solid waste following pyrolysis are two crucial evaluation metrics. Critical effects of pyrolysis parameters on product distribution include those of pyrolysis temperature and pyrolysis time [122]. Reaction temperature and residence time are two crucial variables that affect how effectively pyro-char is produced during the pyrolysis of oily sludge [134].

5.2.5.1. Pyrolysis reaction temperature.

When oily sludge is pyrolyzed, the gas yield increases while the liquid oil and solid residue yields decrease as the pyrolysis temperature rises. Furthermore, too low a temperature prevents complete pyrolysis, resulting in less oil recovery [135]. Higher pyrolysis temperatures allow organics to be converted into non-condensable gases, resulting in gas, liquid, and solid products when the pyrolysis temperature exceeds 800°C, nearly 80 % of the organics convert to gaseous products. Contrary, when the temperature is below 800°C the solid yield increases, at 500°C the maximum oil yield can be around 40% [136]. At the pyrolysis process of oily sludge release H_2S and CH_4S start at 200°C and end at 400°C [137].

5.2.5.2. Pyrolysis reaction time.

Oil recovery effectiveness increases as pyrolysis time increases, peaking at about 60 minutes. Due to further oil degradation into gas and repolymerization into the solid carbonaceous residue, the oil recovery efficiency becomes steady, if not dropping, after that. For this reason, the longer the residence period (60 minutes), the larger the solid yield [138].

5.2.6. Pyrolysis mechanism

Pyrolysis is a thermochemical process that involves the decomposition of organic materials by heat in the absence or near absence of oxygen. This process breaks down complex, long-chain molecules into smaller, simpler ones, producing three main types of products: gases (syngas), liquids (bio-oil or pyrolysis oil), and solids (biochar or char). The mechanisms of pyrolysis are complex and depend heavily on the type of feedstock material, temperature, heating rate, residence time, and the presence of catalysts. General pyrolysis mechanism the overall pyrolysis process typically involves several overlapping stages drying (below ∼100–150°C) [139]. Initial heating removes physically and chemically bound water from the material. This stage is endothermic and consumes significant energy. Devolatilization primary pyrolysis (150–500°C, depending on material) [140]. Regarding the temperature increases, the organic material starts to decompose, releasing volatile compounds, this is the primary stage where most of the organic matter breaks down. This often involves, depolymerization breaking of long polymer chains into smaller fragments. Dehydration removal of water molecules from the molecular structure. Decarboxylation/ Decarboxylation removal of CO_2 and CO groups. Cleavage of C-C and C-H bonds formation of various hydrocarbons, oxygenates, and other volatile compounds. Secondary reactions (higher temperatures, and/or longer residence times) the volatile products released during primary pyrolysis can undergo further reactions if they remain in the hot zone of the reactor [141].

5.3. Microwave process of oily sludge

High frequency electromagnetic waves are generally referred to as microwaves. The temperature distribution and direction of heat transmission for traditional electric heating and microwave heating are shown in Fig. 8. Contrary to microwave heating, which starts from the interior and then transfers heat to the surface of materials, conventional electric heating first heats the surface of materials before transmitting heat to the interior [142]. Oily sludge is heated more quickly and effectively with microwave radiation than with conventional techniques due to molecular interactions with electromagnetic fields [143]. Heating cause demulsification of the oil / water mixture by raising the temperature of the combination, lowering its viscosity, and hastening the settling of water droplets in the mixes [144]. Additionally, the sudden boost in strength has the ability to transform heavy hydrocarbons into lighter ones. Water, the interior component of oily sludge, is more microwave energy absorbent than oil. Water expands as a result of energy being absorbed in the inner phase of the oil/water mixture, lowering the interfacial film and possibly helping separation. The effectiveness of microwave irradiation can be restricted by things like microwave power, microwave duration, and various sludge qualities [145]. When compared to traditional oven and furnace heating methods, the microwave technology has impressive advantages, such as targeted, quick, and energy-efficient heating [146]. The MW technique uses microwave interaction with the

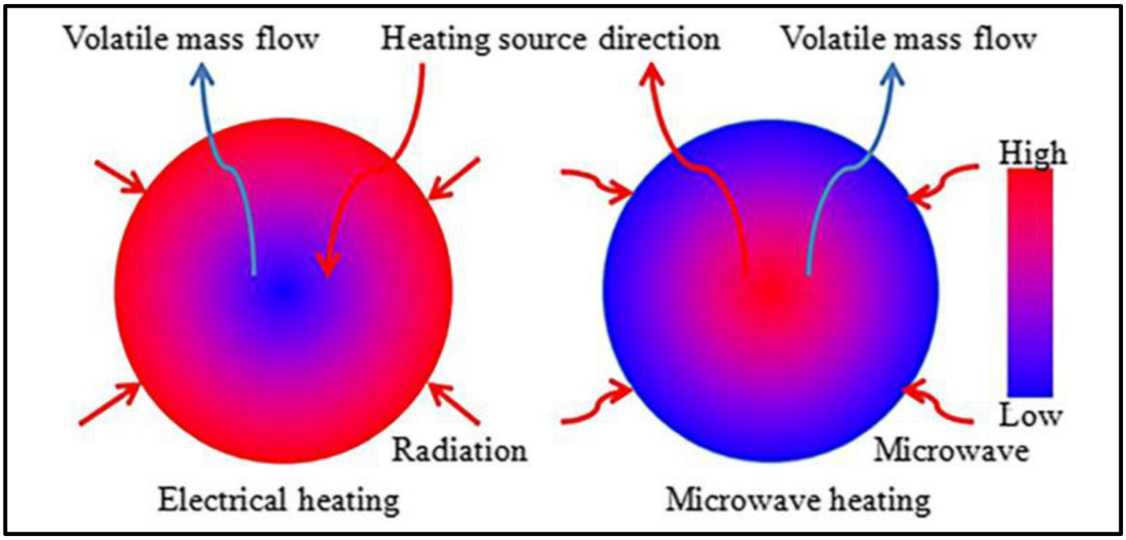

Fig. 8. Conventional heating and microwave heating [147].

Table 17. Microwave processes summarized of oily sludge for micro-char productio.

Oily Sludge Samples	Power W	Micro-char Yield %	HHV MJ/kg	References
China, Zhengzhou, Zhulin AC Co.	600	77.50	34.40	[148]
China, Chongqing Oilfield	800	78.8	–	[149]
China, Shengli oilfield	700	83.2	–	[150]

material's dipoles to generate heat; because this heat is generated from the material's core, there is even heat distribution, great heating precision, and controllability [80]. Table 17 shown the summarized of microwave irradiation processes of oily sludge for micro-char and effect of microwave power on the micro-char yield and HHV recovered.

5.3.1. Effect microwave power

The study by [151] used a range of MW powers for their microwave irradiation technique, including 200, 400, 600, and 800 W. Char yields declined as absorbed microwave power increased, while non-condensable gas yields increased. This suggests that as process temperature rose, more thermal cracking to non-condensable gases occurred.

5.3.2. Effect of irradiation time

Microwave irradiation is a technology for environmentally friendly energy recovery that has a lot of potential for growth in the treatment of oily sludge resources. H. Sun et al, 2019 [152] employed a range of microwave irradiation periods, including 5, 10, 15, 20, and 25 minutes. The MW process, which is commonly used to recover and utilise sludge and other organic wastes, produces high-quality micro-char products.

5.4. Mechanism reaction of thermochemical processes

Mechanism reaction was studied through the characteristics of thermochemical products and reaction kinetics simulation. [153] studies demonstrate that the addition of magnetic (zinc sulphate) particle did not change pyrolysis process of OS. HTC process is an exothermal process that lowers both the oxygen and hydrogen content of the feed described by the molecular (O/C and H/C ratio) [154]. HTC can be defined as combined dehydration and decarboxylation reactions of material to raise its carbon content with the aim of achieving a higher calorific value [110]. The carbon, hydrogen, and nitrogen (CHN) compositions of the char increase as the residence time and reaction temperature were increased [112]. The carbon content increased in the char, while the hydrogen and oxygen contents reduced [113]. These reactions are consistent with the behaviour of feedstock during the HTC process has been attributed to the different reactions such as, dehydration, deoxygenating and decarboxylation reactions that occurred during the HTC process [114]. After the thermochemical process, the H/C and O/C ratios were decreases mainly ascribed to the dehydration and decarboxylation reaction [115]. [98] studies demonstrate the

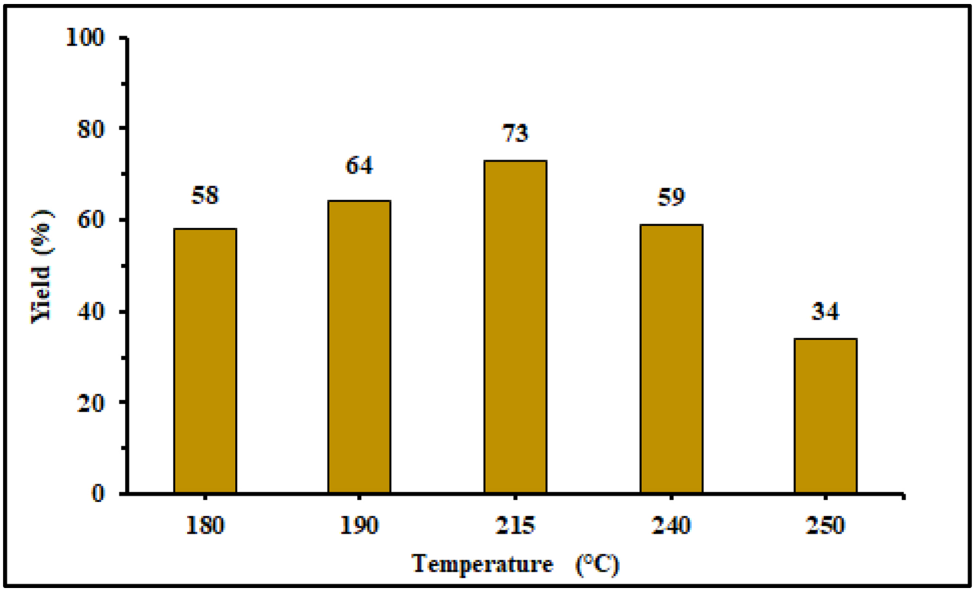

Fig. 9. Char yield with different reaction temperature [162].

mechanism reactions accurse in HTC reaction include decarboxylation, dehydration, condensation, polymerization, aromatization, and hydrolysis. Furthermore, in HTC process main reactions accurse decarboxylation and dehydration [91].

5.4.1. Decarboxylation reaction

The treatment with hydrothermal carbonization allows the carboxyl groups to be partly removed. carboxyl and carbonyl groups degrade rapidly above 150°C, releasing CO_2 and CO respectively. Early studies [98] suggest CO_2; production exceeds predictions based solely on carboxyl reduction. One possible source of CO_2 is formic acid, which is formed in substantial amounts during cellulose degradation and decomposes under hydrothermal conditions mainly to yield H_2O and CO_2 [155]. The term “decarboxylation” usually means replacement of a carboxyl group (-COOH) with a hydrogen atom [156], decarboxylation is one of the oldest known organic reactions [157]. A second technique to remove oxygen from biomass feedstock during the HTC process, producing CO_2 gas, is the decarboxylation reaction. This leads to an increased atomic H/C ratio and a decrease in the atomic O/C ratio, which normally leads to more desirable fuels. Unfortunately, fewer studies on the reaction of decarboxylation relative to the reaction of dehydration have been initiated. Decarboxylation/dehydration were clearly in the results of (FTIR functions groups, and TGA thermal degradation) between [111]. Decarboxylation reaction is one of the processes assumed to accompany pyrolysis and distillation the general formula is:

$RCO_2H \rightarrow RH + CO_2$ [158]

5.4.2. Dehydration reaction

During the HTC process, dehydration reaction may cover both chemical reactions and physical processes which remove water from the feed matrix without altering its chemical composition. Dehydration is called dewatering by lowering the (H/C and O/C) ratios, chemical dehydration significantly carbonizes raw material [98]. A dehydration reaction is a conversion that involves the loss of water from the reacting molecule or ion. Dehydration reactions are common processes, the reverse of a hydration reaction [157]. Common dehydrating agents used in organic synthesis include sulfuric acid and alumina [159]. Often the dehydration reactions are affected by heating [160].

$RCO_2H + R'OH \rightleftharpoons RCO_2R' + H_2O$ [158]

5.5. Energy characteristics of chars

5.5.1. Char yield (%)

The thermochemical conversion process is used for the production of char from raw materials [161]. Characterization of char is done using the following steps to evaluate the char fuel type; comparative studies of the energy characteristics, including product yield (%), calorific value (HHV), proximate, element composition, H/C and O/C atomic ratios of chars. Fig. 9 shows solid yields for the HTC reaction temperature condition. The yields of hydrochar produced

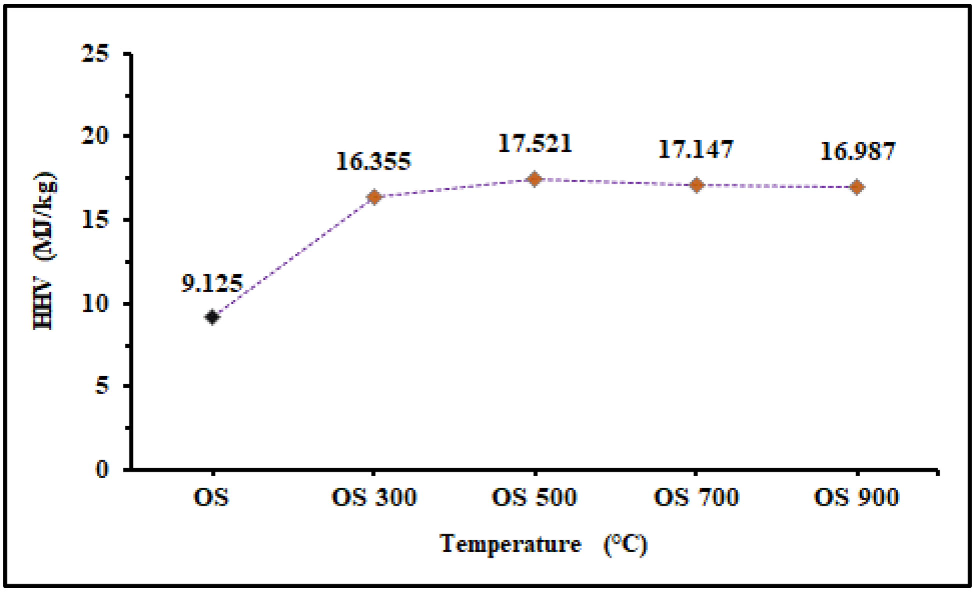

Fig. 10. Effects of reaction temperature on hhv of char [125].

were found to be between 34 and 73%. Although higher treatment temperatures and longer residence times were anticipated to result in lower hydrochar yields, the results show that the HTC parameters taken into account in this literature review had no effect on the change in mass yields. Fig. 9 show the yield percentage as reported by [162]. The solid product is inversely proportional to temperature, as seen in Fig. 9. In the HTC process, the solid product reduces as the temperature rises till 200°C.

5.5.2. Calorific value

In order to assess the potential value of hydrothermal carbon as a solid fuel, the high heating value HHV was established. Fig. 10 demonstrates how char HHV is affected as a function of the parameters of hydrolysis temperature [163]. The char HHV increases from 9.125 MJ/kg to 16.355 MJ/kg as the hydrolysis storage temperature increase to 900°C [125]. As shown in Fig. 10, the reaction temperature can effectively boost char HHV; for instance, the char HHV at 900°C increased 53%, the char HHV was increasing to 500°C, then began to decrease. Furthermore, high temperatures will result in more energy being used. The char HHV suggests that the impact as a fuel has been accomplished when compared to the raw material HHV [164].

5.5.3. Proximate and ultimate analysis of chars

The proximate results of the char product derived from the thermochemical conversion process can be read in different ways and according to what is available in the research laboratories, such as the European Standards (EN ISO 18134e2) estimated by [165]. The study by [166] studies demonstrate proximate analysis using a different method determined according to EN 15403:201 and EN 15402:2011 for ash and volatile matter, respectively. The char was subjected to proximate and ultimate analysis using the American Standards for Research and Materials (ASTM) methods and the Vario E1 Cube elemental analyser (Elementar Co. Ltd, Germany), respectively [167]. Table 17 shows the proximate analysis of chars produced by thermochemical conversion processes. Table 18 shows the ultimate analysis of chars produced by thermochemical conversion processes. As s results in Tables 18 and 19, we can see a discrepancy in the values of the approximate and ultimate analysis, which indicates that the oily sludge is a complex compound. The results also indicate that oily sludge is usually collected randomly in storage yards exposed to sunlight [168].

5.5.4. Van Kervelen diagram

Van Krevelen diagrams are charts designed by Dirk Willem van Krevelen, a scientist and professor of fuel technology at TU Delft, and used to ascertain the formation and maturity of kerogen and petroleum. The diagram plots the oxygen: carbon atomic ratio as a function of the hydrogen: carbon atomic ratio [169, 170]. The Van Krevelen diagram was used to represent hydrochar evolution [171]. This diagram shows the chemical effect on hydrochar product through the influence of dehydration and decarbonization reactions [154, 156, 172]. studies demonstrate [173] that the hydrolysis technique permits the determination

Table 18. Proximate analysis of chars.

		Proximate analysis				
Oily sludge Samples	processes	M %	V %	Ash %	FC %	Reference
China, Nanyang oil field	HTC @ 210 °C	–	58.73	39.97	1.30	[106]
China, Nanyang oil field	HTC @ 240 °C	–	61.41	37.95	0.64	[106]
China, Nanyang oil field	HTC @ 270 °C	–	61.29	38.6	0.11	[106]
Iraq, NRC Baiji Alsomod	PY @300 °C	–	30.8	39.7	29.5	[125]
Iraq, NRC Baiji Alsomod	PY @500 °C	–	30.5	41.2	29.3	[125]
Iraq, NRC Baiji Alsomod	PY @700 °C	–	30.3	40.1	29.6	[125]
Chennai, Manali, oilfield	MW @ 450 W		13.1	12.7	74.2	[74]
Chennai, Manali, oilfield	MW @ 600 W		17.7	14.4	67.9	[74]
Chennai, Manali, oilfield	MW @ 800 W		12.0	13.3	74.7	[74]

Table 19. Ultimate analysis of chars.

		Ultimate analysis					
Oily Sludge Samples	Processes	C %	H %	N %	O %	S %	Reference
India, petroleum refinery	HTC @ 150°C	54.82	5.31	0.31	34.59	4.97	[73]
India, petroleum refinery	HTC @ 175°C	56.42	5.03	0.29	33.31	4.95	[73]
India, petroleum refinery	HTC @ 200°C	36.86	4.56	0.27	26.97	4.34	[73]
Iraq, NRC Baiji Alsomod	PY @300 °C	44.98	1.30	1.12	52.60	0.0	[125]
Iraq, NRC Baiji Alsomod	PY @500 °C	31.93	1.20	1.21	65.62	0.0	[125]
Iraq, NRC Baiji Alsomod	PY @700 °C	38.15	1.5	1.24	59.10	0.0	[125]
Chennai, Manali, oilfield	MW @ 450 W	69.4	1.2	0.5	11.2	5.0	[74]
Chennai, Manali, oilfield	MW @ 600 W	75.2	0.9	0.4	6.5	3.5	[74]
Chennai, Manali, oilfield	MW @ 800 W	79.0	0.8	0.4	2.0	4.5	[74]

of the whole spectrum of parameters that describes the qualitative and quantitative characteristics of the sludge, including total organic carbon (TOC), oxygen and hydrogen indices (OI and HI), maximum hydrocarbon yield temperature at full hydrolysis maximum temperature. The associated Van Krevelen diagram for hydrolytic parameters (HI and OI) shows the original properties of the organic matter. The Van Krevelen diagram offers the opportunity not only to identify kerogenous fields of different types of hydrochar but also to follow their evolution.

6. Review summary

There has been a lot of interest in figuring out alternate ways to deal with the vast amount of oily sludge that is produced in the oil sector. Although oily sludge is now underutilized, it is carbon-rich and has a huge potential for the manufacture of fuel. Even if it has been completely remedied, oily sludge cannot be dumped in a landfill [174]. Using the HTC technique, oily sludge has been converted to solid carbon in a few trials. According to a previous study, HTC offers a viable way to turn waste and low-quality materials into materials that are similar to energy-dense coal. There is no requirement for pre-drying because HTC is a wet process and oil sludge contains between 30 and 50 percent water.

The process of pyrolysis involves the anaerobic breakdown of oily sludge at temperatures between (200 and 800°C) to produce a very dense energy pyro-char. The oily sludge is chemically destroyed utilizing thermochemical methods. Similar to this, in conventional pyrolysis, the energy from the power source is transformed into heat and then transferred to the surface and subsequently the interior sections of the target material via convection and conduction [175]. Similar to this, the surface temperature and thermophysical properties of the selected material, such as heat capacity, density, and thermal conductivity, frequently limit heating performance [176]. The microwave technique uses microwave irradiation with powers between 200 and 800 W. Oily sludge is heated using the microwave approach, which is a relatively novel heating technique [177]. The microwave heating system has many benefits over conventional oven and furnace heating systems, including targeted, quick, and energy-efficient heating. By using microwave interaction with the selected material's dipoles to generate heat, the microwave technique produces heat from the material's core, resulting in controllability, equal heat distribution, and high heating precision [80].

NRC Baiji produces about 3000 to 3500 cubic meters of oily sludge annually that is not previously treated, which led to the accumulation of very large quantities estimated at thousands of tons; these quantities caused obstacles and confusion in the company's work and are stored randomly and irregularly. In Baiji, about 90% of oily sludge is discharged to landfill in the Makhoul Mountains Baiji which causes adverse effects on land. In addition to the damage, it causes harm to the environment and humans due to its toxicity [25].

The actual need for fuel in the NRC Baiji to operate the production units, refinery units, and support units such as energy units and other administrative units is high. These issues prompted ideas on finding alternative methods of energy generation, such as the HTC, pyrolysis, and, microwave processes which has good advantages such as low operating temperature, short time, small area, and the production of carbon-rich fuel with a high calorific value and low CO_2 emission rate. The char will be used as an alternative fuel, which rid the environment of accumulated pollutants and produces low-cost fuel. This research will bring about increase in the financial profits of the NRC Baiji. This study will investigate the possibility of converting oily sludge to solid carbon fuel char; find the percentage solid carbon in oily sludge with best conditions achievable in the HTC, pyrolysis, and microwave processes. In addition, the disadvantage of the methods of treating oily sludge currently have been summarized take for instance.

Regarding current treatment methods, the study [178] studies demonstrate the solvent extraction in the filed application, recovered 70% of oil has many disadvantages are large scales extraction, low efficiency and high variability. [179] centrifugation method applied in field recovered 50% of oil disadvantages of this process are large space for the installation of the plant, it is very costly and pose environmental concern. Surfactant enhanced oil recovery (EOR) applied in field 80% of oil recovery, while it has flaws large space for the installation of plant, very costly, pose environmental concern as a study [180]. The method of freeze/ thaw in the laboratory work recovered around 60% of oil retrieval has disadvantages of high temperature, long duration time, High energy consumption[181]. Electro kinetic method in the lab rotaries experiment has recovered 60% oil recovered the disadvantages complicated, application, only in small scale [182]. Ultrasonic irradiation process in the laboratory application was reached 70 of oil recovered the disadvantages are very costly [183]. Froth flotation laboratory method has recovered oil reached 60% has low efficiency [184]. Incineration in the field was recovered about 90 while disadvantages are high cost of equipment, environmental pollution [185]. Stabilization solidification the laboratory teste about 90% oil recovered disadvantage of this process is only for oily sludge with low moisture content (or dry state) [186]. Oxidation of laboratory application has reached around 90 of oil recovered the disadvantage are high cost of operation, environmental pollution [187]. Land farming field 80% oil recovered had many disadvantages are slow process, require a very large portion of land and can pose environmental concerns. Biopile composting in the field 80% had disadvantages high cost of operation and require land area [31]. Bio slurry field about 90% oil recovered but it has disadvantages high cost of operation and proper management of the end product [188].

7. Conclusions

In this review oily sludge properties and thermochemical conversion processes (HTC, PY, and MW) were detailed. This work showed the explanation of the literature review by comprehensive and detailed described the sources of oily sludge, its nature and risk level to humans and the environment. It provides deep understanding of the previous treatment methods for the recovery of oil and energy from oily sludge using different methods, as well as the disadvantages of each method which led to the research gap. Depending on the objectives of this study, this work focused on the study of the physicochemical property of OS, determined ability it undergo thermal chemical conversion processes. Three concerns are the main obstacles in the chars produced from OS using HTC, pyrolysis, and microwave, respectively. Firstly, understanding the chemical processes that take place throughout the conversion process is more difficult for HTC than turning sludge into products, which is necessary for turning oily sludge treatments into highly effective and cost-effective solutions. Therefore, the optimum conditions were found (200°C, 600 min for HTC of OS, as well as 600°C, 60 min for PY of OS, while 800 W, 15 min for MW of OS). Quantitative reaction models based on dominant reaction pathways are necessary for reactor design and process optimization. Second, accurate estimation and setup are necessary to implement the integrated process combined with pyrolysis and the energy consumption of thermal treatment for oily sludge; as well as examining the distribution of dangerous substances in pyrolytic by-products and figuring out how to pyrolyze oily sludge to release pollutants and change them. Finally, the microwave process is more energy-efficient than other heating methods because microwave irradiation may quickly increase the energy of the medium's molecules, leading to faster reaction rates in a short amount of time. However, due to equipment requirements and high running costs, its industrial application is constrained.

Conflict of interest

The author declares no conflict interest.

Ethical approval

Not applicable.

Data availability

The data will be available upon request.

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Abbreviations

OS	Oily sludge
WOS	Waste oily sludge
PAHs	Polycyclic aromatic hydrocarbons
HTC	Hydrothermal carbonization
TH	Thermal hydrolysis
HTL	Hydrothermal liquefaction
PHWE	Pressurized hot water extraction
WAO	Wet air oxidation
SCWU	Supercritical water upgrading
SCWO	Supercritical water oxidation
SCWG	Supercritical water gasification
W/O	Water in oil
O/W	Oil in water
PHCs	Polyaromatic hydrocarbons
ZN	Zinc
Cu	Copper
Ni	Nickel
Pb	Lead
Cr	Chromium
Hg	Mercury
NSO	Nitrogen, sulphur, and oxygen
N	Nitrogen
S	Sulphur
O	Oxygen
NRC	North Refineries Company
TSR	Total solid residue
TOC	Total organic carbon
PY	Pyrolysis process
PAH	Polycyclic aromatic hydrocarbon
AHC	Aliphatic hydrocarbon compounds
DRO	Diesel range organic
GRO	Gasoline range organic
ORO	Oil range organic
HHV	High heating value
M	Moisture
VM	Volatile matter
FC	Fixed carbon
C	Carbone
H	Hydrogen
N	Nitrogen
S	Sulphur
O	Oxygen
CO_2	Carbon dioxide
MPa	Mega pascal
HTG	Hydrothermal gasification
CP	Conventional pyrolysis
MWP	Microwave pyrolysis
PY	Pyrolysis
MW	Microwave
O/C	Oxygen / carbon
H/C	Hydrogen / carbon
TOC	total organic carbon
OI	Oxygen indices
HI	Hydrogen indices
EOR	Enhanced oil recovery

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