Review of petroleum sludge thermal treatment and utilization of ash as a construction material, a way to environmental sustainability

Mubarak Usman Kankia 1, Lavania Baloo 1,*, Bashar S. Mohammed 1, Suhaimi B. Hassan 2, Effa Affiana Ishak 1, Zakariyya Uba Zango 3

1Department of Civil and Environmental Engineering, Universiti Teknologi Petronas, Seri Iskandar, Malaysia
2Department of Mechanical Engineering, Universiti Teknologi Petronas, Seri Iskandar, Malaysia
3Department of Fundamental and Applied Sciences, Universiti Teknologi Petronas, Seri Iskandar, Malaysia

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A B S T R A C T

Oil companies are largely responsible for producing a huge amount of petroleum sludge (PS), which is a major source of pollution in the environment. It is generated during oil extraction, processing, transportation, and cleaning processes. Environment Protection Act and Hazardous Wastes Handling Rules categorized petroleum sludge as hazardous waste because it consists of spent chemicals, wastewater, waste oil, mineral matter, and contaminated sand. This PS cannot be disposed of in a landfill, even after it is effectively de-oiled. However, PS treatment and disposal are serious threats for most refineries. Thus, the treatment became crucial. In this paper, a comprehensive review of PS sources, characteristics, environmental effects along with the comparative analyses of available thermal and disposal methods of PS treatment are presented. This review paper could enhance the essential knowledge and future guide for PS thermal and disposal techniques.

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1. Introduction

In the present day, petroleum is one of the fundamental sources of generating energy (Deng et al., 2015). Raw oil extraction and refining process continue to increase due to the rise in demand by automobile companies, metal plating equipment, mining processes, fertilizer companies, batteries, tanneries, paper industries, and pesticide companies in many countries (Xiao et al., 2019). Thus, with the fast economic development of the world, energy remains the main life-blood for economic growth (Hu et al., 2017). Therefore, petrochemical companies produce different types of effluents and wastes during crude oil refining, installations, and cleaning processes. Petroleum production worldwide is about 12,600,000 m³ per day, while around 190,000 m³/day of petroleum sludge (PS) is generated (Ramirez and Collins, 2018). Thus, 1.51% of the production. PS is a viscous, and thick combination of waste oil products, solid residue, water, and hydrocarbons which is generated from the process of raw oil refining, storage and vessel cleaning (Ramirez and Collins, 2018; Fitri et al., 2017; Nazem and Tavakoli, 2017). The origin source and petroleum types are the fundamental factors that govern the PS complexity (Wang et al., 2015). The mixture of petroleum sludge is produced mainly as a result of sedimentation and accumulation in storage tanks of raw petroleum, transportation tanks/pipelines (Wang et al., 2012). PS contains petroleum hydrocarbons such as aromatic, aliphatic, asphaltene, and nitrogen sulfur-oxygen components (Nazem and Tavakoli, 2017; Aguelmous et al., 2018). Oily sludge is characterized by alkanes 40–52%, aromatics 28–31%, resins 7–22.5%, and asphaltenes 8–10% (Lin et al., 2018). It consists of 30–40% water, 10–20% solid particles, and 30–80% oil by mass (Lin et al., 2017). PS is generally considered as a toxic waste due to the existence of harmful compounds, solid particles, heavy metals, water/oil emulsion and persistent and recalcitrant sediments (Aguelmous et al., 2018; Lin et al., 2018; 2017; Shen et al., 2016). Currently, among the major challenges and global problems for humanity are environmental pollution and energy insecurity (Ghaleb et al., 2020). The major areas of producing petroleum sludge are raw oil storage vessels and separation of water from oil (Ramirez and Collins, 2018). Other sources of petroleum sludge are from operating slop, American
Petroleum Institute (API) separator bottom, operating residue, and oil spill (Wang et al., 2015).

Effective PS management is significantly required. Reuse and recycle wastes are the ideal opportunities for sustainable development (Jagaba et al., 2019). There are different techniques for the treatment of petroleum sludge, and the choice depends on the desired aim to achieve (Ramirez and Collins, 2018). The removal methods of oil from the petroleum sludge can be done by froth flotation, surfactant EOR (enhanced oil recovery), microwave irradiation, ultrasonic radiation, solvent extraction, freeze/thaw, centrifugation, pyrolysis and electro-kinetic (Hu et al., 2017; Nazem and Tavakoli, 2017; Shen et al., 2016; Silva et al., 2019; Pazoki and Hasanidarabadi, 2017). These methods may either partially or totally decrease the harmful substances in petroleum sludge to a minimum acceptable level (Gong et al., 2017). Among the available techniques, the promising process is pyrolysis, which decreases the toxic content such as heavy metals and polycyclic aromatic hydrocarbons (PAH), besides that, it does not cause any air pollution problem during the process (Lin et al., 2019).

In addition, the final disposal of PS can be achieved by bio-slurry, landfill, land farming, composting bio-pile, stabilization and solidification (encapsulation), and incineration methods (Nazem and Tavakoli, 2017; Silva et al., 2019). Some of the methods used for petroleum sludge disposal have shown to be effective, while some proved to have high capital cost (installation of equipment), require very high equipment maintenance and some may lead to secondary environmental problems, for example, the emission of harmful gases (incineration method) (Silva et al., 2019; Pazoki and Hasanidarabadi, 2017).

Researchers gave attention to the pyrolysis method because of its high recovery of energy potential and a relatively small amount of pollutant discharge (Lin et al., 2018). Pyrolysis is defined as the thermal disintegration of organic substances at an elevated temperature in a free-oxygen domain and oil recovery from petroleum sludge pyrolysis obtained between 460 – 650°C (Lin et al., 2018; Cheng et al., 2017). Pyrolysis reaction is divided into two steps, which are primary (endothermic reactions) and secondary (exothermic reactions) pyrolysis (Pánek et al., 2014). Comparing the pyrolysis method with other available treatments, the pyrolysis method recovers a large quantity of oil and better crude material flexibility and versatility that make it a standout amongst the best choices for the treatment of petroleum sludge and recovery of oil (Cheng et al., 2017). Catalyst (additive), heating rate, oxygen content, and temperature are the fundamental factors that affect petroleum sludge pyrolysis products (Lin et al., 2018). The change of petroleum sludge into different valuable materials, for example, atomic weight, organic substances, and carbonaceous sediment by utilization of pyrolysis tackle the disposal issue as well as the resources management (Pánek et al., 2014). The by-products of pyrolysis are low molecular weight hydrocarbons in liquid form, along with incondensable gases and char or ash (Hu et al., 2013). Pyrolysis is used widely for the manufacture of oil from different biomass streams, for instance, cotton stalk and rice husk (Lin et al., 2018).

Stabilization/solidification (S/S) is a successful and efficient process of disposal that can limit the effects of these harmful substances in an area (Xiao et al., 2019). Stabilization-solidification is a fast and cheap oily-sludge treatment strategy; desired to deactivate hazardous contaminants by changing over them into a less dissolvable or a less harmful form (i.e., stabilization), in addition, by encapsulating the stabilized contaminants into a matrix form, i.e., solidification (Hu et al., 2013). In the method of encapsulation, binder (cement) is used to encapsulate or seal the toxic substances in an inactive and closed space by the physicochemical process. This method gives maximum qualities, for example, anti-soaking, mechanical properties, anti-drying, and anti-leaching properties (Xiao et al., 2019). Pozzolan is usually added to enhance the solidification performance.

In most construction, the concrete is used very widely as a construction material. It is composed of four fundamental components, which are water, cement, coarse and fine aggregates. These four basic ingredients are proportioned correctly and homogeneously mixed together to produce concrete. In some cases, an admixture or pozzolanic additive is required to increase the desired performance of certain physical and mechanical properties of concrete. The additives are added with the aim of raising the concrete hardness, density, durability, corrosion resistance, compressive and flexural strength (Boikova et al., 2017). These concrete properties largely depend upon the proper selection of water/cement ratio (w/c ratio), properties of coarse and fine aggregate, and thorough concrete compaction during casting in the formwork (Nagrockiê et al., 2017).

Concrete as a construction material offers significant benefits; among them, it gives a chance either to adjust its ingredients fully or partially to manufacture a very strong and durable concrete that meets the standard characteristics. In some instances, cement has been substituted either partially or fully by some materials like slag, pumice powder, fly ash, silica fume, cow bone ash, stainless steel, waste of agricultural products such as rice husk ash, coconut shell ash, palm kernel shell ash (Rubio-Cintas et al., 2019; Nguyen et al., 2019; Kabay et al., 2015). On the other hand, fine and coarse aggregates were replaced either partially or fully with the materials mentioned before to manufacture lightweight concrete or high-performance concrete (Utsev and Taku, 2012).

In this review, petroleum sludge sources, characteristics, and effects of direct disposal to the soil were discussed. The pyrolysis process is deemed to be an effective method that reduces the toxicity and conversion of PS to three useful products, which
are pyrolysis oil, ash, and syngas. With consideration of sustainability, the ash could be turned into a valuable product, instead of disposing of in the landfill. Thus, to treat the PS, convert the ash into the usable product as cement replacement material in concrete. Moreover, to ensure the ash is disposed of in a safe way, encapsulating the ash in concrete is anticipated as an environmentally friendly approach, and significantly reduces the leaching of the harmful content in the ash.

2. Sources, characteristics, and toxicity of petroleum sludge

2.1. Sources of petroleum sludge

In previous decades, crude oil extraction has continued to increase rapidly. The transportation and collection of crude oil inevitably result in a huge volume of oily waste in the tanks of oil storage (Xiao et al., 2019). The different exercises engaged with the oil industry, for example, boring, generating, and transporting, make some hazardous waste (Silva et al., 2012). Both activities at the upstream and downstream in the oil industry can produce a lot of oily sludge. The upstream task incorporates the procedures of extricating, transporting, and putting away raw petroleum, while the downstream activity involves the refining of raw petroleum. The produced oily sludge in the oil industry is grouped as either simple oil or oil waste upon the proportion of solid materials and water inside the oil residue (Hu et al., 2013). The petroleum sludge has a high volume of solid residues, very viscous in nature, while simple oil has less percentage of water (Hu et al., 2013). Petroleum sludge, the most hazardous waste created in petroleum refineries, semisolid, pasty material made of sand (a clay blend, silica, and oxides) tainted by oil, the water created, and the additives (chemicals) utilized in oil manufacturing (Silva et al., 2012). The oily sludge generated in the process of extraction of crude oil at the upstream and in the refining process at the downstream activity is summarized in the Fig. 1 (Hu et al., 2013; Khalil et al., 2018; Islam, 2015).

Specifically, the base residue in unrefined petroleum storage vessels is the most seriously studied oil waste in wide literature. When crude oil is extracted, it is stored firstly in storage tanks. In the vessels, the heavier substances of petroleum hydrocarbons have the ability to settle down together with solid residues and water (Hu et al., 2013). Heavier hydrocarbons (C_{20+} hydrocarbon molecules) separate from lighter hydrocarbons (Heath et al., 2004). The oily sludge made up of water, oil, and water deposited in a vessel base (Fitri et al., 2017). The oil waste evacuated in the cleaning process and forwarded for further necessary actions (treatment and disposal). Unrefined petroleum properties (viscosity and density), method of refining, the most significant is the capacity of

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**Fig. 1:** A scheme showing the sources of petroleum sludge
refining, the mode of storage is affecting oil waste generation (Hu et al., 2013). Generally, the oil-processing industry can produce a tremendous quantity of oily waste estimated in the range of 3–5kg for every ton of treated raw petroleum (Aguelmous et al., 2018).

A research carried out by US EPA, every oil processing plant in the United States, generates yearly about 30,000 tons of petroleum sludge. In China, the yearly generation of petroleum sludge from the processing industry of crude oil is assessed to be three million tons. In general, the oily sludge production depends heavily on the petroleum industry’s refining capacity. It has been evaluated that 1 ton of petroleum sludge is produced in every 500 tons of raw petroleum prepared (Hu et al., 2013). In the refining process of 1kg crude oil, petroleum sludge of 10–20 grams can be produced. Most transported and unrefined oils have a separation tendency. This issue is frequently exacerbated by the venting of unpredictable segments from the unrefined oil, cool temperatures, and the static state of liquid during stockpiling. The substantial particles separate from the unrefined petroleum and accumulated in the bottoms of tanks are called “tank bottoms” or “oily sludge” (Heath et al., 2004).

2.2. Petroleum sludge characteristics

Oil or raw petroleum is normally happening combustible fluid comprising of a complex blend of hydrocarbons of different compounds of liquid and other molecular weights, which are obtained in geologic arrangements underneath the earth’s surface (Islam, 2015). Crude oil contains hydrocarbons such as aromatic hydrocarbons-phenol, polyaromatic hydrocarbons (PAHs), BTEX-benzene, toluene, ethylbenzene, and xylene, besides alkanes, organic compounds (sulfur, oxygen, and nitrogen), inorganic compounds (suspended solids, water-soluble metals, and salts), cycloalkanes and some alike vanadium, iron, copper and nickel. However, from one formation to another, there is a wide variation in molecular composition (Islam, 2015; Coca et al., 2011; Reddy et al., 2011).

The petroleum sludge composition is very difficult to understand and deal with (Islam, 2015). It involves suspended solids and oil in water, water in oil emulsion (Abdurahman and Yunus, 2006). Petroleum sludge includes dangerous substances such as poly cyclic aromatic hydrocarbons, aromatic hydrocarbon (Young and Cerniglia, 1995), and complete hydrocarbons content (Ayotamuno et al., 2007). Petroleum sludge is hard to hydrate because of its high thickness (viscosity). Petroleum sludge is a harmful waste. The sludge essentially contains about 55.13% of water, 9.246% of residues, 1.9173% of asphaltenes, 23.19% of light hydrocarbons, and 10.514% of wax. Likewise, high metals concentrations, for example, Fe is 0.6%, vanadium is 204 ppm, and nickel is 506ppm, which makes petroleum waste very destructive for the earth and creatures, which should be managed for ecological protection (Islam, 2015).

Oily sludge chemical composition differs from one petroleum sludge to another, thus largely depends on the source of the raw oil (oil field), the process of drilling refining (Silva et al., 2019). Petroleum sludge is characterized by high oil hydrocarbons content range between 5–86.2% (Aguelmous et al., 2018; Khalil et al., 2018). The petroleum sludge pH value normally ranges between 6.5 and 7.5 (Hu et al., 2013). Typical oily sludge is a complex mixture that contains a considerable amount of different substances such as water, oil, hydrocarbons, several toxic/poisonous, carcinogenic, or mutagenic compounds (Huang et al., 2017; Wang et al., 2018a). In general, petroleum sludge consists of 40–52% alkanes, 8–10% asphaltenes, 28–31% aromatic hydrocarbons, and 7–22.4% of resins (Aguelmous et al., 2018).

2.3. Petroleum sludge toxicity

Oil processing plants fractionate the unrefined petroleum overall by thermal cracking to acquire condensed oil gas (liquefied petroleum gas-LPG), lamp fuel (kerosene), diesel oil, naphtha, and other leftover fuel oil. However, these industries are not environment-friendly, releasing harmful compounds, for example, toluene, benzene, and xylene in the magnitude of 2.5g emitted per ton of refined petroleum. In addition, the emission of volatile organic substances ranging from 0.5 to 6kg/t of unrefined petroleum (Aguelmous et al., 2018). Because of its pollution characteristics, in Europe and China, the oily sludge has been categorized as hazardous waste (Wang et al., 2018a).

Oily sludge is considered as unsafe waste due to the existence of harmful compounds, solids, heavy metals, water-in-oil emulsion, and other sediments (Aguelmous et al., 2018). Because of the presence of harmful substances high concentration, the inappropriate method of disposal of petroleum sludge may result in serious environmental threats; when the surrounding soil receives the sludge, both chemical and physical properties of the soil are affected, leading to change in soil morphology (Robertson et al., 2007). The petroleum sludge polluted soils can make a deficiency of nutrients and hinder seed germination, and cause limited development or kill plants on contact (Al-Mutairi et al., 2008). The soil pores may be fixed by the sludge components because of their high viscosity (Trofimov and Rozanova, 2003). This could lead to a decrease in soil hygroscopic moisture, soil ability to retain water, and soil hydraulic conductivity (Trofimov and Rozanova, 2003; Suleimanov et al., 2005). The heavy metals and petroleum hydrocarbons (PHCs) present in oily sludge may cause different toxic effects to be human and ecological receptors (Robertson et al., 2007). Moreover, PHCs can penetrate through soil layers to the groundwater and resulting adverse negative effects on the soil enzymes, microorganisms, and
aquatic animals (Trofimov and Rozanova, 2003; Wake, 2005).

2.4. Hazardous waste

As a result of fast economic and industrial development, a considerable amount of hazardous waste is produced as a by-product nowadays. Naturally, hazardous waste possesses at least one of the ignitibility, corrosivity, reactivity, and toxicity characteristics (Arimoro et al., 2008; Johnson et al., 2014). Hazardous waste has the potential negative effects on health and the environment (Johnson et al., 2018; Ma et al., 2019). Drying is a major process of hazardous waste treatment (Wake, 2005; Sotoudehniakarani et al., 2019). Pyrolysis can also be pretreated prior to the process of disposal are more expensive, time-consuming, and resulting in minor contamination (Hu et al., 2017). The integrated thermal method of treatment for petroleum sludge is a labor-saving procedure for waste treatment (Gong et al., 2018). The thermal disintegration process of petroleum sludge is a promising treatment method for the effective utilization of the hazardous petroleum sludge (Deng et al., 2015), because of its volatile organic materials and high heating value (Ma et al., 2019). Pyrolysis has been the most cost-effective, efficient, and acceptable thermochemical technique for converting hazardous waste materials to useful energy products (Tang et al., 2018; Campuzano et al., 2019). The resulted hydrocarbons from the process of pyrolysis are of lesser molecular weight (Sotoudehniakarani et al., 2019).

### Table 1: Acceptable contaminants concentration for the toxicity characteristic

| EPA HW No. | Contaminant | Regulatory Level (mg/L) | EPA HW No. | Contaminant | Regulatory Level (mg/L) |
|------------|-------------|-------------------------|------------|-------------|-------------------------|
| D004       | Arsenic     | 5.0                     | D032       | Hexachlorobenzene | 0.13                   |
| D005       | Barium      | 100.0                   | D033       | Hexachlorobutadiene | 0.5                  |
| D018       | Benzene     | 0.5                     | D034       | Hexachloroethane | 3.0                  |
| D006       | Cadmium     | 1.0                     | D008       | Lead        | 5.0                  |
| D019       | Carbon tetrachloride | 0.5 | D013       | Lindane    | 0.4                  |
| D020       | Chloride    | 0.03                    | D009       | Mercury     | 0.2                  |
| D021       | Chlorobenzene | 100.0         | D014       | Methoxychlor | 1.0                  |
| D022       | Chloroform  | 6.0                     | D035       | Methyl ethyl ketone | 200.00          |
| D007       | Chromium    | 5.0                     | D036       | Nitrobenzene | 2.0                  |
| D023       | o-Cresol    | 4 200.0                 | D037       | Pentachlorophenol | 100.00          |
| D024       | m-Cresol    | 4 200.0                 | D038       | Pyridine    | 5.0                  |
| D025       | p-Cresol    | 420.0                   | D010       | Selenium    | 1.0                  |
| D026       | Cresol      | 1 200.0                 | D011       | Silver      | 5.0                  |
| D016       | 2,4-D       | 10.0                    | D039       | Tetrachloroethylene | 0.7                |
| D027       | 1,4-Dichlorobenzene | 7.5 | D015       | Toxaphene  | 0.5                  |
| D028       | 1,2-Dichloroethane | 0.5       | D040       | Trichloroethylene | 0.5                |
| D029       | 1,1-Dichloroethylene | 0.7      | D041       | 2,4,5-Trichlorophenol | 400.00          |
| D030       | 2,4-Dinitrotoluene | 3 0.13  | D042       | 2,4,6-Trichlorophenol | 2.0                |
| D012       | Endrin      | 0.02                    | D017       | 2,4,5-TP (Silvex) | 1.0                |
| D031       | Heptachlor (and its epoxide) | 0.008      | D043       | Vinyl chloride | 0.2                |

1. Hazardous waste number; Chemical abstracts service number; 2. Quantitation limits are greater than the calculated regulatory level. The quantitation limit, therefore, becomes the regulatory level. 3. If o-, m-, and p-Cresol concentrations cannot be differentiated, the total cresol (D026) concentration is used. The regulatory level of total cresol is 200mg/L.

3. Pyrolysis technique

3.1. Petroleum sludge pyrolysis

The pyrolysis method simply means the disintegration of natural materials thermally at high temperatures in an inert domain (500–1000°C) (Hu et al., 2013; Johnson et al., 2018). Pyrolysis can also be defined as the procedure of thermochemical disintegration of organic compounds at a higher temperature and without oxygen to create a solid product (ash/char), liquid (pyrolysis-oil), as well as gases such as CO, H₂, CH₄, and CO₂ (Sotoudehniakarani et al., 2019; Wang et al., 2018b; Liu et al., 2009). It is said to be an endothermic reaction.

Petroleum sludge is a mixture that contains more than the water of about 50% of its total weight. Thus, it requires to be pretreated prior to the process of pyrolysis (Gong et al., 2017). Drying is a major pretreatment process that highly decreases the water content, raises the oily sludge calorific value, declines the cost of storage and transport, increases the combustion rate (Deng et al., 2015). Distinctively, petroleum tank bottom sludge can be utilized through microbial degradation, in addition, utilization into valuable oils. However, it was discovered that such processes pose secondary pollution (Liu et al., 2009). And, the methods of final disposal are more expensive, time-consuming, and resulting in minor contamination (Hu et al., 2017). The integrated thermal method of treatment for petroleum sludge is a labor-saving procedure for waste treatment (Gong et al., 2018). The thermal disintegration process of petroleum sludge is a promising treatment method for the effective utilization of the hazardous petroleum sludge (Deng et al., 2015), because of its volatile organic materials and high heating value (Ma et al., 2019). Pyrolysis has been the most cost-effective, efficient, and acceptable thermochemical technique for converting hazardous waste materials to useful energy products (Tang et al., 2018; Campuzano et al., 2019). The resulted hydrocarbons from the process of pyrolysis are of lesser molecular weight (Sotoudehniakarani et al., 2019).
al., 2019; Wang et al., 2018b; Campuzano et al., 2019). The petroleum sludge char may be used as a source of fuels in power plants (Wang et al., 2018b). Liu et al. (2009) stated that it is of paramount importance to do pyrolysis kinetic studies so as to have sufficient knowledge about reaction rate, decomposition mechanisms, and reaction parameters and anticipate the distribution of the products. Moreover, these studies could help to select the proper reactor, reactor design enhancement, and the conditions of operation.

A research performed by Schmidt and Kaminsky (2001) as well as Deng et al. (2015) on petroleum sludge pyrolysis using fluidized bed and temperature range of 460–650°C observed 70–84% of oil could be recovered from the oily sludge. The effective recovery of oil from petroleum sludge occurs between 460–650°C with the maximum oil yield of 70–80% (Johnson et al., 2018). Moreover, Hu et al. (2013) stated that the optimum yield of hydrocarbons in oily sludge pyrolysis was recorded at the temperature of 440°C. The major disintegration of petroleum sludge occurred at a temperature range of 100–350°C whereas the inorganic components started breaking down when the heating temperature reached 400°C and the carbon content in the solid residue, at the end of the pyrolysis temperature (900°C), recorded as 38wt.% of the initial petroleum sludge (Karayildirim et al., 2006).

Johnson et al. (2018) and Arazo et al. (2017) reported that bottom tank oily sludge pyrolysis led to a rise in retrieving rate of oil at elevated temperature (525°C), however, when the heating was more than 525°C the oil yield decreased because of the reactions of secondary composition which may lead to falling out of gaseous and lighter hydrocarbons from the oil. Also, researchers (Deng et al., 2015; Hu et al., 2013; Chang et al., 2000; Silva et al., 2017) found that the reaction of pyrolysis for oily sludge began from a low temperature of about 200°C (473K). In addition, they reported that the optimum evolution rate had been noted from 350–500°C (623–773K) using the thermogravimetry-mass spectrum (TG/MS). During the pyrolysis process, total organic carbon (TOC) of about 80% may be converted to valuable hydrocarbons, from 327–450°C temperature.

Gong et al. (2018) conducted research on the pyrolysis of petroleum sludge. They discovered from thermogravimetry (TG), and differential thermogravimetric (DTG) curves that pyrolysis of took place when the temperature reached 200°C to the optimum temperature of about 700°C. In addition, as the pyrolysis temperature increased, the yield of the char declined while the gas production raised. However, oil yield declined when the temperature was above 600°C, and the char percentage was 55% recorded. Thermogravimetric analysis (TGA) is a weight loss determination method with the aid of temperature increase. The data of TGA may be used for structural component analysis (proximate analysis), thermal stability checks, kinetic reaction formulation, and analysis of materials. Fig. 2 shows the potential reaction route during co-pyrolysis of petroleum sludge with rice husk at 600°C.

![Fig. 2: Potential reaction route during co-pyrolysis of petroleum sludge with rice husk at 600°C (Lin et al., 2018)](image-url)
Lin et al. (2018) conducted the co-pyrolysis of petroleum sludge, and rice husk was carried out in a fixed bed reactor to study the interaction effects on the products as well as to enhance bio-oil quality. Fig. 2 depicts the feasible reaction track of the pyrolysis process of petroleum sludge, and rice husk was incorporated. The rice husk was burnt prior to 400°C (Wang et al., 2013a). Moreover, the solid residue was produced when the heavy substances of petroleum sludge began to decompose. The petroleum sludge volatiles could interact as well as accumulate on the char, resulting in an increase of the solid residues yield. In addition, the synergy led to a decrease in the oil yield and an increase in the syngas yield. This might be associated with the synergetic effect of alkali metal provided by the biomass ash (rice husk), hence, enhancing the secondary reactions of hydrocarbons such as dehydrogenation and cracking.

The kinetics studies of petroleum sludge pyrolysis were recently examined using thermogravimetric analysis (TGA), and model fitting was mostly employed to predict the pyrolysis process (Chang et al., 2000; Shie et al., 2000; 2002; 2003; 2004a). Several studies present the model-fitting method used for isothermal data that yields explicit figures of Arrhenius parameters, which are probably to cover multi-step kinetics; no isothermal data continually result in extremely unclear kinetic triplets. Therefore, an appropriate model fitting technique is critically required for the utilization as well as the interpretation of the kinetic triplets. Several integrals are isoconversional techniques such as Flynn–Wall–Ozawa equation (FWO), Coats–Redfern equation, Popescu method, and Kissinger–Akahira Sunose equation accept that the standards of $E_a$ and ‘a’ remains constant in the reaction until the required level of conversion (α) is achieved, making the techniques slightly comparable to the inflexible global one-step models that accept an unchangeable $E_a$ for pyrolysis methods (Burnham and Dinh, 2007). If $E_a$ depends upon α, thus, it was revealed that the integral is conversion technique usage might result in systematic errors (Zhou et al., 2017). FWO technique can directly calculate the $E_a$ related to other methods, and it can sidestep the error from dissimilar assumptions of the reaction mechanism function.

Typically, the kinetic reactions of thermal decomposition are presented by the nth-order reaction equation (García et al., 2001; Wang et al., 2013b; Lefebvre et al., 2003) (Eq. 1 to 3). In addition, the Arrhenius equation (Eq. 4) is usually used to explain the reaction rate constant.

$$\frac{da}{dt} = K(T)f(\alpha)$$  
$$\alpha = (W_i - W(t)/W_i - W_f) \times 100\% \rightarrow 0\% - 100\%$$  
$$f(\alpha) = (1 - \alpha)^n$$  
$$K(T) = Aexp(-Ea/RT)$$

where $T$ and $t$ are the temperature and operating time; $da/dt$ is the relationship between the rapid conversion ratio and the operating time, and $\alpha$ is the conversion ratio. From Eq. 2, $Wi, Wt, and Wf$ are designated as initial weight, weight at a time, and final weight of the reaction, respectively. From Eq. 1 and 4, $K(T)$ is the constant reaction rate. Furthermore, from Eq. 3, $f(\alpha)$ is the nth-order rate of reaction equation; $n$ is the reaction order. Similarly, in Eq. 4, $A$ is the pre-exponential factor; $R$ is the gas constant; and $Ea$ is the activation energy.

The focal objective for thermal kinetic analysis is to acquire three basic elements (Jiang et al., 2018), i.e., the pre-exponential factor (A), the representation of the nth-order reaction rate function ($f(\alpha)$), and the activation energy ($Ea$). Clearly, it is inaccurate to differentiate the mass signal against temperature or time in a single non-isothermal or isothermal thermogravimetric test. Hence, multiple non-isothermal thermogravimetric analyses are frequently practical for thermal kinetic studies. If the associations among the reaction time and operating temperature are in the form of Eq. 5, and for the meantime, the heating rate was constant in a certain TGA test, Eq. 1 could be re-written as Eq. 6:

$$T = \beta t + To \Rightarrow dt/dT = \beta$$  
$$da/dT = A/B \times \exp(-Ea/RT) f(\alpha)$$

where $To$ and $T$ are the initial temperature and operating temperature respectively; $\beta$ is the heating rate; $da/dT$ is the relationship between the instantaneous conversion ratio and the operating temperature. However, Eq. 6 is the basic differential formula for the investigation of thermal kinetic analysis that represents the relationship between the instantaneous conversion ratio of the reactant with $T$ under specific heating rate ($\beta$). The Coats–Redfern method (Coats and Redfern, 1964) and Friedman (1964) method were obtained through rearranging Eq. 6, and it was effectively applied for the thermal kinetic analysis in the pyrolysis technique of petroleum sludge (Shie et al., 2000). However, differential methods have some existing limitations and are mainly due to $da/dT$, which was intensely affected by the background noise of the TGA test (Lim et al., 2016; Font and Garrido, 2018; Orava and Greer, 2015). Thus, the activation energy ($Ea$) acquired in differential methods was inaccurate. According to Eq. 6, the integral method for the study of thermal kinetic analysis could be concluded as follows:

$$\frac{da}{dT} = -\frac{A}{\beta} \exp(-Ea/RT) f(\alpha) \Rightarrow 1/f(\alpha) \frac{da}{dT} = -\frac{A}{\beta} e^{-Ea/RT} dT$$  
$$G(\alpha) = \int_0^T 1/f(\alpha) \frac{da}{dT} = A/\beta \int_{To}^T e^{-Ea/RT} dT$$

where $G(\alpha)$ is an integral equation of $f(\alpha)$; $To$ and $T$ are the initial and final operating temperatures, respectively.

The pre-exponential factor (A) and activation energy ($Ea$) can be obtained via rewriting Eq. 8, and the negative effect of the background noise could be ignored. However, the solution of the nth-order
reaction rate equation \( f(\alpha) \) and reaction order \((n)\) were difficult to get. Thus, both the integral method and differential method and have their limitations and advantages during the attainment of the pre-exponential factor, activation energy, and the nth-order reaction rate equation. 

Punnaruttanakun et al. (2003) studied thermal and kinetic characteristics, using different heating rates, of API separator oily sludge pyrolysis by thermalgravimetric analysis (TGA). Shie et al. (2000; 2002; 2003; 2004a; 2004b) and Chang et al. (2000) have conducted several kinds of research on the petroleum sludge pyrolysis; examined the main by-product produced from petroleum oil sludge pyrolysis with the addition of catalysts by TGA. Research by Gong et al. (2018) examined the pyrolysis of petroleum sludge with the addition of aluminum, iron, sodium, potassium, and solid waste. Moreover, they observed that the catalytic additives had many effects on the pyrolysis of petroleum sludge. The pyrolysis of oily sludge was enhanced and proper utilization of the by-products (oil, char, and gases). Silva et al. (2017) did their research on acid as a catalyst additive and temperature effect on pyrolysis and the recovery of oil. They found that the catalytic and thermal impacts on the pyrolysis could be possibly utilized to convert the petroleum sludge to a fraction of diesel oil. Fig. 3 illustrates the principle of the pyrolysis process of petroleum sludge in an oxygen-free environment.

![Diagram of pyrolysis process](image)

**Fig. 3:** A diagram showing the process of pyrolysis (Campuzano et al., 2019)

### 3.2. Pyrolysis fundamentals

Several pyrolysis literature reviews can be obtained (Roy and Dias, 2017; Peacocke and Bridgwater, 2000). The reviews encompass reactors of pyrolysis, the process parameters of pyrolysis, and the pyrolysis products. From the literature, pyrolysis may be classified as conventional, flash, or fast, depending upon the process parameters for the operation employed. The conventional pyrolysis is also known as slow pyrolysis (Mohan et al., 2006), refers to thermal disintegration of organic substances in a free oxygen environment at a relatively lower rate of heating (0.1–10°C/s), whereas fast pyrolysis is associated with a process of high-temperature (400–550°C) in which the feedstock is quickly heated (10–200°C/s) and broke down to produce char, pyrolysis oil, and gases. The liquid oil is obtained by quick quenching as well as cooling the vapors. While flash pyrolysis is also a thermal decomposition process of a feedstock at an extremely high rate of heating greater than 1000°C/s along with lower vapor residence time so as to decrease the secondary cracking, which results in high yield of liquid (Pokorna et al., 2009; Akhtar and Amin, 2012).

The typical researched area of pyrolysis is the core aspect of the reactor, although the improvement and control of the pyrolysis products are receiving more attention. In the literature, different reactors were used to pyrolyze different industrial by-products such as petroleum sludge, including circulating fluidized bed reactors (Zhou et al., 2009), fluidized bed reactors, fixed bed reactors (Liu et al., 2009), rotary kilns, batch or semi-batch reactors as well as other innovative solutions such as solar or plasma (Bridgwater, 1999; Uluisik et al., 2017; Butler et al., 2011).
reactors for the pyrolysis process largely depend on the intended process (Marshall et al., 2014). Distinctively, the equipment of pyrolysis consists of a cyclone, reactor along with a condenser. The cyclone removes the char from the liquid and gas (Matayeva et al., 2019). Fig. 4 depicts the pyrolysis process of petroleum sludge. Table 2 provides an overview of the reactors, process parameters as well as classification of pyrolysis.

![Pyrolysis Diagram](image)

**Table 2: An overview of the pyrolysis types, process parameters, and reactors**

| Type of pyrolysis | Pyrolysis temperature (°C) | Heating rate (°C/s) | Particle size (mm) | Vapor residence time | Suitable reactors | Sources |
|-------------------|---------------------------|---------------------|-------------------|---------------------|-------------------|---------|
| Fast              | 500-700                   | 10-200              | <1-2              | 0.5-10 s            | Ablative, auger, fluidized bed, circulating fluidized bed reactors | (Marshall et al., 2014; Matayeva et al., 2019; Fonts et al., 2012; Guda et al., 2015; Krutof and Hawboldt, 2016) |
| Flash             | 500-1000                  | >1000               | <0.2              | <1 s                | Fluidized bed, circulating fluidized bed reactors, downer reactors | (Marshall et al., 2014; Matayeva et al., 2019; Moharir et al., 2019) |
| Slow              | 300-500                   | 0.1-10              | 5-50              | 5-30 min            | Fixed bed, vacuum reactors | (Mohan et al., 2006; Marshall et al., 2014; Matayeva et al., 2019; Moharir et al., 2019; Zhang and Matharu, 2018) |

4. Stabilization and solidification

Stabilization and solidification techniques are typically methods which comprise a waste mixing with the addition of a binder to decrease contaminant migration ability via both chemical and physical means as well as to transform the harmful waste into a waste form that is environmentally acceptable for construction use or land disposal.

The stabilization-solidification method can be defined as the measure of sealing or encapsulating hazardous wastes (petroleum sludge char) with the aid of binding materials so as to prevent harmful substances leaking into the surrounding (Johnson et al., 2018). Portland cement has been used as a binding material widely for deactivation and encapsulation method of heavy metals so as to save costs and energy (Karamalidis and Voudrias, 2008; Li et al., 2014). Hu et al. (2013) defined stabilization and solidification as a cost-effective and fast hazardous waste treatment method aimed to render the hazardous contaminants inactive by reducing their toxicity to the lowest acceptable level (i.e., stabilization/chemical fixation) while solidifying/encapsulating them into a matrix form of maximum structural integrity (Hu et al., 2013). In addition, encapsulation can be achieved through chemical or physical means and to have a successful
conversion of petroleum sludge char to eco-friendly building material (Johnson et al., 2018). Stabilization is a hazardous waste treatment using chemical agents like phosphoric acid or lime to decline mobility of the harmful contaminants, while Solidification refers to less permeability confinement, and this enhances mechanical properties. Moreover, it reduces the hazardous waste surface of contact with the leaching elements (Rubio-Cintas et al., 2019).

The use of cement to immobilize and encapsulate hazardous waste comprises three main stages, which are:

- Rectifying chemical contaminants: this involves interactions chemically between the contaminants and cement products hydration.
- Adsorbing present contaminants physically on the hydrated cement product.
- The confinement of hazardous waste.

In the phases, first and second above, it is important to make a good observation of the hazardous waste and hydration products behavior, because they are the ascertaining factors for results, whereas the third phase relies upon the kind of the product of hydration and the type pore structure and paste characteristics (Johnson et al., 2018). A research conducted by Karamalidis and Voudrias (2007) on oily sludge encapsulation using Portland cement, and they discovered a concentration increase of leaching metals due to cement content increase.

For the purpose of enhancing the immobilization performance of the stabilization and solidification method, it is of paramount importance to incorporate admixtures (pozzolanic substances) in the process. Thus, the pozzolanic materials raise the sorption ability of the organic compound. Caldwell et al. (1990) conducted an experiment with encapsulation. They used Portland cement with the incorporation of activated carbon as a pozzolan to deactivate organic contaminants, and there was a positive increase in the result. However, activated carbon is very expensive. An alternative and affordable material that exhibits binding and adsorption characteristics should be used. Leonard and Stegemann (2010) discovered that the addition of high carbon fly ash (HCFA) to Portland cement in the stabilization and solidification of oily sludge waste as an adsorbent and the result revealed that HCFA effectively declined the leachability of petroleum hydrocarbons. During the petroleum sludge treatment process, concrete samples were cast in molds and placed in airtight plastic bags to avoid carbonation because of air exposure, and the curing duration was 24 hours in a humid chamber with a temperature of about 21± 3°C. Prior to mold dismantling, the samples were being recoated with the plastic-bags for a further cure in a humid chamber for 7, 28 and 56 days before testing time.

Zain et al. (2010) conducted research and reported that 5%, 10%, and 15% incorporation of rice husk ash in the stabilization and solidification petroleum sludge contaminants as a partial cement replacement. Thus, the obtained results of the compressive strength of the matrix are between 19.2 and 24.9(N/mm²). Al-Futaisi et al. (2007) used Portland cement (OPC), and other pozzolanic admixtures quarry fines (QF), cement by-pass-dust (CBPD) to solidify bottom tank petroleum sludge. They carried out the toxicity characteristic leaching procedure (TCLP), and the result indicated that all the values of heavy metals are within the permissible TCLP values of metals (Zain et al, 2010).

5. Conclusion

Petroleum sludge is a hazardous waste, and its direct disposal poses threats to the environment and human health. The disposal is banned in most countries by regulatory bodies like the resource conservation and recovery act (RCRA) and the US Environmental Protection Agency (US EPA). Pyrolysis is a promising process that reduces the contaminants to acceptable levels according to regulatory standards. It is also an inexpensive technology that is simple to operate and process a large variety of biomass. The technique of pyrolysis decreases a considerable volume of wastes going to disposal areas as well as greenhouse gas emissions. It brings water pollution risk to the lowest level. The pyrolysis method provides jobs for low-income earners. The ash contains a considerable amount of heavy metals, which may cause environmental pollution. In addition, it has pozzolanic characteristics like fly ash, rice husk ash and can be used as a construction material. Stabilization chemically fixes or alters the hazardous waste by deactivating them while the solidification process confines the deactivated waste into a form of a matrix.

Compliance with ethical standards

Conflict of interest

The authors declare that they have no conflict of interest.

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