Treatment of Wastewater from Petroleum Industry: Current Practices and Perspectives

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Abstract

Petroleum industry is one of the fastest growing industry and it significantly contributes to economic growth in developing countries like India. The wastewater from a petroleum industry consist a wide variety of pollutants like petroleum hydrocarbons, mercaptans, oil and grease, phenol, ammonia, sulphide and other organic compounds etc. All these compounds are present as very complex form in discharged water of petroleum industry, which are harmful for environment directly or indirectly. Some of the techniques used to treat oily waste/wastewater are membrane technology, photocatalytic degradation, advanced oxidation process, electrochemical catalysis, etc. In this review paper we aim to discuss past and present scenario of using various treatment technologies for treatment of petroleum industry waste/wastewater. The treatment of petroleum industry wastewater involves physical, chemical and biological processes. This review also provides scientific literature on knowledge gaps and future research directions to evaluate the effect(s) of various treatment technologies available.

Key words: Petroleum hydrocarbons; Membrane technology; Photocatalytic degradation; Waste biorefinery; Resource recovery

1) Introduction

Water is necessary for life on earth as it is a basic need of all organisms. Rapid industrialization and economic development have led to exponential growth in population and urbanization (Zafra et al. 2015; Chen 2018). The world is witnessing an increase in urbanization and industrialization due to the consumerist approach (Li and Yu 2011; Zhang et al. 2015). The domestic and industrial sectors continuously generate large amount of waste/wastewater at an alarming rate and usually dispose the waste without proper management and treatment (Varjani et al. 2019). Petroleum industries and refineries are important from economic growth point of view (Li and Yu 2011; Li et al. 2014; Abdulredha et al. 2018). Petroleum refineries and petrochemical plants are facing problem of disposing the waste/wastewater generated. Wastewater released by petroleum industries contains different type of organic and inorganic pollutants such as BTEX, sulphides, hydrocarbons, phenol, heavy metals etc (He and Jiang 2008; Usman et al. 2012; Varjani 2017a; Raza et al. 2018). Large quantities of toxic substances are released by activities of petroleum industry such as production process of oil, transportation, oil refinery, petrochemical product, storage and distribution etc. These are harmful for environment and human health (Perera et al. 2012; Viggi et al. 2015; Raza et al. 2018).

The treatment of oily wastewater generated from petroleum industry involves different processes such as (1) physical, (b) chemical and (c) biological processes Various processes available for treatment of petroleum hydrocarbons polluted water ranges from applied methods to emerging methods (Usman et al. 2012; Li et al. 2014; Viggi et al. 2015; Varjani 2017b). Characteristics of oily wastewater and effect of petroleum hydrocarbons on environment and human health have been emphasized in this paper. Sections of this paper also discusses about the current treatment process used in petroleum industry and treatment methods used for treating oily wastewater. The current review focuses to generate critical review about the different practices used for wastewater treatment in petroleum industries. This review also focusses on information pertaining to the knowledge gaps and future research directions in this field.

2.) Characteristics of oily wastewater

Petroleum means “rock oil” and this word is derived from a Latin word “petra” and Greek word “oleum” (Jafarinejad 2016). Petroleum industry generates large amount of oily waste either solid or liquid due to upstream and downstream operations (Varjani and Upasani 2017). Upstream process includes extracting, transporting and storing crude oil and downstream process includes refining of crude oil (Al-Futaisi et al. 2007; Hu et al. 2013; Thakur et al. 2018). Depending on a ratio of water and solids waste is categorized into simple wastewater crude oil or sludge. The pH value of oily sludge is usually ranging from 6.5 to 7.5, but it varies depending on sources of crude oil, processing method, reagents used etc. (Hu et al. 2013; Jasmine and Mukherji 2015). Wastewater from petroleum industry contain oil impurities, high level of BOD and COD, high total solids, hydrocarbons and other waste. This waste includes oily sludge, waste catalyst, heavy metals, volatile organic compounds, oil & grease content, high total dissolved salts, ammonia, nitrates, sulfides, etc. (Jasmine and Mukherji 2015). Oily wastewater contains mainly four major types of petroleum hydrocarbons such as aliphatic, aromatic, asphaltenes and compounds containing oxygen, nitrogen and sulfur. It consists organo-metallic complexes with cadmium, lead, nickel and vanadium (Honse et al. 2012; Varjani and Upasani 2017; Thakur et al. 2018). Table 1 summarizes the characteristics of oily wastewater/effluents and standards for their discharge in environment. These pollutants typically disperse, emulsify or dissolve within the oily wastewater. In general aromatic and aliphatic compounds counts up to 75% of petroleum hydrocarbons in oily wastewater (Ward et al. 2003; Perera et al. 2012; Jasmine and Mukherji 2015; Varjani et al. 2018).

3) Effect on environment/ human health

Petroleum industry discharge huge amount of pollutants in environment. Wastewater released by the petroleum industries contain large quantities of hydrocarbons, heavy metals, phenols and other toxic chemicals (Perera et al. 2012; Jasmine and Mukherji 2015; Thakur et al. 2018; Varjani et al. 2018). Different activities of petroleum industry such as transportation, storage, drilling etc. take part in soil contamination by oil. Depending on type of soil lighter oil can move quickly instead of heavier oil through the soil layers (Fakhru'l-Razi et al. 2009; Varjani et al. 2017). Due to ineffectiveness of treatment system, industrial wastewater become harmful to ecosystem and other life forms (Poulopoulos et al. 2005; Veyrand et al. 2013; Varjani 2017b; Al-Hawash et al. 2018). Oily wastewater can affect different components of environment such as human health, drinking water and groundwater resources, air, crop production and aquatic life etc (Zafra et al 2015; Varjani et al. 2017). Accumulation of toxic products in the water bodies leads serious consequences on the ecosystem and living organisms either long term or short term which may be chronic or acute. (Poulopoulos et al. 2005; Usman et al. 2012; Al-Hawash et al. 2018; Varjani et al. 2018).

4) Current treatment process in petroleum industry

Different technologies adsorption, coagulation, anaerobic treatment, reverse osmosis, ultrafiltration, chemical destabilization, flocculation, dissolved air flotation (DAF), membrane process etc. have been used to treat wastewater from petroleum industry (Bennett and Peters 1988; Kriipsalu et al. 2003; Sonune and Ghate 2004; Usman et al. 2012; Li et al. 2014; Padaki et al. 2015). Adsorption method has many advantages rather than other technique such as cost benefit, simplicity and adaptability (Sabah et al. 2007; Ahmad et al. 2016). Depending upon concentration and source of contamination different type of treatment technique is required to reduce the toxic level of pollutants (Sonune and Ghate 2004; Hanafy and Nabih 2007; Bennett and Peters 1988; Farajnezhad and Gharbani 2012; Li et al. 2014; Padaki et al. 2015). Mainly the treatment process has been differentiated into three categories (a) primary, (b) secondary and (c) tertiary treatment (fig.1). Generally effluent of petroleum industry is passed through different stages for reducing toxicity level which is show as a schematic diagram in in fig. 2.

4.1) Primary treatment

Primary treatment is usually used for physical operation in petroleum refinery wastewater treatment plant (Ahmad et al. 2016). It is important step as it allows waste for the further secondary treatment unit. Mostly gravity separation applies to classify the floating and settle down material from wastewater. The primary treatment step includes an oil/water separator which can separate oil, water and solids. Gravity separation followed by skimming is carried out for removal of oil from wastewater (Al-Shamrani et al. 2002; Hanafy and Nabih 2007). Oil-water separator such as API oil–water separator is widely used because of low cost, high effectiveness for primary treatment step. API separators work on phenomena of difference in specific gravity to allow heavy material to settle below lighter liquids. Hydrocarbons that float on the surface and the sludge settle down to the bottom (Al-Futaisi et al. 2007; Ahmad et al. 2016). API separator is not very much applicable for removal of smaller oil droplets and emulsions (Abdulredha et al. 2018). Dissolved air flotation (DAF) is a water treatment process that uses air to increase the capacity of smaller oil droplets and enhance the separation process. DAF unit typically consist chemicals to promote coagulation and increase floc size to make easy separation. In this stage the heterogeneous components of the effluent such as suspended solids colloids or dispersion, immiscible liquids are reduced significantly (Al-Shamrani et al. 2002; Hanafy and Nabih 2007). Colloids and dispersion also delay and damage equipment during proceeding stage (Renault et al. 2009). In Induced air flotation (IAF) system, air is induced by rotor disperse mechanism, the spinning rotor work as a pump and forces to the fluid. The advantages of the IAF process are compact size, low cost and effective removal of free oil and suspended material (Bennett and Peters 1988; Bennett and Shammas 2010).

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4.2) Secondary treatment

The main purpose of this stage is to reduce contamination level of effluent and make it in permissible limit for discharge into water bodies. Secondary treatment consists coagulation, flocculation and further biological treatment to reduce toxicity of petroleum wastewater (Xu and Zhu 2004; Viggi et al. 2015; Changmai et al. 2017). Petroleum effluent contains large number of refractory components. Poly aluminium chloride is more effective rather than ferric chloride in coagulation process for treatment of petroleum wastewater (Farajnezhad and Gharbani. 2012; Hosny et al. 2016). Coagulation–flocculation is a process in which chemical product is added to accelerate the sedimentation in clarification tank ~~Coagulation-flocculation consist on the addition of chemical product to accelerate the sedimentation in clarification tank~~ (Kriipsalu et al. 2003; Moulai-Mostefa and Tir, 2004, Hosny et al. 2016). The coagulants are organic or inorganic components such as aluminium hydroxide chloride, aluminium sulphate or high molecular weight cationic polymer. The aim of addition of coagulant is to remove 90% of the suspended solids from the wastewater (Lin and Wen 2003; Changmai et al. 2017). Renault et al. (2009) have reported chitosan for efficient coagulation/flocculation process to treat petroleum industry wastewater. (Renault et al. 2009). Biological treatment is the most widely used method for removal of organic compounds in the oil industry wastewater. Biological treatment is mostly classified in to two categories (a) suspended growth process and (b) attached growth process (Chavan and Mukherji 2008; Srikanth et al. 2018). Suspended growth process includes aerated lagoon, membrane bioreactor technology, sequencing batch reactor (SBR) and activated sludge treatment. In the activated sludge process the wastewater enters in to aeration tank where microorganism brought in contact with contaminated wastewater. Microorganisms use the organic material as food and decompose organic matter (Srikanth et al. 2018). N:P ratio has been reported very important parameter for treatment of oily wastewater by using oil degrading bacteria (Chavan and Mukherji 2008).

4.3) Tertiary treatment

Tertiary treatment process includes sand filtration, activated carbon process and chemical oxidation. It is applicable for removal of total suspended solids, dissolved and suspended matter, COD and trace organics such as PAHs (He and Jiang 2008; Li et al. 2014). After secondary treatment process, effluent contain suspended solids depending on operating conditions in the clarifier. Removal of metals and fine solids which could not be settle down in sedimentation process can be removed by sand filtration (Zahrim and Hilal 2013; Ahmad et al. 2016). This process involves passing the wastewater through a filter bed comprised of a filter media. Generally chemical oxidation is used for reduction of residual COD, trace organic compounds and non-biodegradable compounds. This method uses different type of oxidation reagents like hydrogen peroxide, ozone, chlorine dioxide (Usman et al. 2012; Srikanth et al. 2016).

5) Treatment methods for oily wastewater

It is important to dispose of effluent in a proper manner and it should maintain a minimum concentration level of chemical(s) which is suitable for environment. Thus, innovative research is required to develop new technologies which degrade the complex molecules into simpler forms, which is reliable to maintain water quality (Sonune and Ghate 2004; Ani et al. 2018). Petroleum industries have made few effective technologies for improving treatment capacity through different methods which are mentioned below and also have also been summarized in table 2.

5.1) Membrane separation technology

Membrane separation processes are dependent on difference in (a) pressure, (b) concentration of pollutants, (c) temperature and (d) electrical potential (Al-Obaidani et al. 2008; Jamaly et al. 2015; Adhama et al. 2018). Depending upon the pore size of utilized membrane process is categorized as a (a) microfiltration (MF), (b) nanofiltration (NF), (c) ultrafiltration (UF) and (d) reverse osmosis (RO), which are mostly applied to treat oily wastewater (Bruggen et al. 2003; Tomaszewska 2007; Jamaly et al. 2015). This separation method play role in physical removal of way of the trapped particle size of contaminants (Hilal et al. 2004). According to membrane pore size ultrafiltration membranes are more effective than microfiltration membranes. Nanofiltration and reverse osmosis can be also used for separating oil from water especially for high salinity water (Zhu et al. 2014; Jamaly et al. 2015). Based on the oil dispersion, oily water can be classified into three types: (a) free-floating oil, (b) unstable oil-water emulsions and (c) stable oil-water emulsions (Srijaroonrat et al. 1999, Hilal et al. 2004; Qiu et al. 2009). Generally free-floating oil can be easily removed by mechanical techniques and also unstable emulsions removed mechanically but sometimes specific chemical additives are required (Salahi et al. 2013; Abdulredha et al. 2018). Membrane separation efficiency is normally identified by oil rejection coefficient (Ro) and it is defined as:

$$\mathbf{R_0 = \frac{Oil\ concentration\ in\ feed - Oil\ concentration\ in\ permeate}{Oil\ concentration\ in\ feed} \times 100}$$

Usually, an effective membrane has high rejection coefficients for total organic carbon (TOC), total surface charge (TSC) and chemical oxygen demand (COD) (Karhu et al. 2013). Another important parameter in membrane separation is the permeate flux. Permeate flux is defined by:

$$\mathbf{J = \frac{Vp}{At}}$$

Where **J** is the permeate flux, **Vp** is the permeate volume, **A** is the membrane effective area, and **t** is the permeation time (Rezvanpour et al. 2009). The permeate flux also depends on membrane properties such as porosity, pore size and hydrophilicity (Mohammadi et al. 2003; Changmai et al. 2017). Due to small pore size of membrane, oil rejection coefficient is higher and use of large pore size membrane leads high permeate flux (Kocherginsky et al. 2003; Colle et al. 2009; Han et al. 2017).

5.2) Advanced oxidation process

The main function of advanced oxidation process is to generate high reactive free radicals. Hydroxyl radicals are effective in destroying organic chemicals because they are reactive electrophiles (Oller et al. 2011; Jamaly et al. 2015). Hydroxyl radical is strong oxidant to destroy compound that cannot be oxidized by conventional oxidant. Hydroxyl radical possess faster oxidation rate as compared to H_2O_2 or $KMnO_4$ (Gogate and Pandit 2004; Usman et al 2012; Tijani et al. 2014). Generated hydroxyl radicals can attack organic chemicals by radical addition, hydrogen abstraction and/or electron transfer (Gogate and Pandit 2004; Tijani et al. 2014). Generation of hydroxyl radical is commonly accelerated by combining O_3, H_2O_2, TiO_2, UV radiation, electron-beam irradiation and ultrasound (Usman et al 2012; Li et al. 2014; Varjani 2017b).

5.3) Electrochemical catalysis

Electrochemical catalysis process is related to oxidation of hydroxyl radical with a highly organic matter between substitution, addition and electron transfer process (Zhang et al. 2015; Mohan et al. 2018). It leads to the degradation of pollutants, mineralization, easy to build airtight circulation with no secondary pollution (Lin et al. 2001; Dimoglo et al. 2004; Koper 2005; Bajracharya et al. 2015; Changmai et al. 2017). Electrolysis process is applied to treat oily wastewater which leads to time dependent reduction in chemical oxygen demand (COD). The process consists three steps for effective reduction of pollutants in oily wastewater and do the remediation. The first step is the direct oxidation of oil components at the electrode, by the metal oxide itself or by hydroxyl radicals available at electrode surface. The second step is indirect oxidation of oil components by intermediate oxidizing agents formed and the third step is aggregation of suspended oil droplets by electro-flotation (Santos et al. 2006; Jamaly et al. 2015). Electrochemical catalytic treatment is more effective treatment of oily wastewater (Mohan et al. 2018). Ma and Wang (2006) have performed research with pilot scale plant having double anodes and cathodes. Anode contained active metal and graphite, however cathode contained iron and a noble metal content catalyst with big surface. They have reported approximately 90% reduction of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) and 99% reduction in suspended solids by using the electrochemical process.

5.4) Photocatalytic Degradation

Photolytic degradation is very well researched in last decade and conventionally a method for treating petroleum wastewater (Twesme et al. 2006; Do et al. 2010; Yen et al. 2011; Varjani 2017b). Photocatalysis is beneficial process used to oxidize persistent compounds which cannot be oxidized during biological treatment (Vodyanitskii et al. 2016). TiO_2 and ZnO are commonly used photocatalysts in the treatment of petroleum industry wastewater (Santos et al. 2006; Akpan and Hameed 2009; Vodyanitskii et al. 2016). Benefits of TiO_2/UV technique are low cost, faster reaction rates, no sludge production and easy operation at ambient temperature and pressure with complete mineralization of petroleum industry wastewater (Twesme et al. 2006; Akpan and Hameed 2009; Varjani 2017b). Park and Choi (2005) have reported photocatalytic hydroxylation of aromatic ring in presence of platinum loaded TiO_2, where they have used water as an oxidant. Photocatalytic degradation of naphthalene present in petroleum industry waste using TiO_2 in presence of inorganic anions had been reported by Lair et al. (2007).

6) Knowledge gaps and Future perspectives

Petroleum hydrocarbon pollutants are classified as priority pollutants. The pollutants which are present in petroleum industry wastewater can be effectively remediated using different technologies (Yen et al. 2011; Vodyanitskii et al. 2016). Research on study of innovative technologies with minimum environmental and economical influence indicating that it is thrust areas of research (Lin et al. 2001; Santos et al. 2006; Viggi et al. 2015; Vodyanitskii et al. 2016). This review article focuses on improving different technologies used for treatment of wastewater generated by petroleum industry activities. Petroleum industry wastewater contain different toxic substances such as toluene, xylene, benzene, ethylbenzene, phenols and PAHs, etc (Hanafy and Nabih 2007; Renault et al. 2009; Varjani et al. 2017). These pollutants are very difficult to be directly removed by using single method specifically biological treatment which is considered as green approach. Hence advanced chemical or physical treatments in combination with biological treatment are required. Apart from this for biological treatment to identify the successful species which can work *ex-situ* and *in-situ* conditions is a very important task (Varjani 2017b; Chen 2018). It has been reported that integration of various processes may give better results than individual process used for treatment of oily wastewater. But knowledge of integration of technologies is still at its infancy which needs to be explored by researchers. Recent developments are mostly more expensive, require maintenance and it's a time taking process. Hence, all industries do not participate to reduce toxicity of effluent. Future technology needs easy operating systems which are suitable for small, medium and big industries.

Huge amount of solid and liquid waste is produced due to petroleum industry activities. Management and handling of waste generated is nowadays is a big issue for local authorities not only in urban areas i.e. municipalities but also other regions in any country (Santos et al. 2006; Li and Yu 2011; Tijani et al. 2014; Jamaly et al. 2015; Varjani and Sudha 2018). Due to increased urbanization and industrialization generation of wastewater, appropriate disposal, treatment and/or recycling is posing more challenges as treatment and disposal costs large amount in terms of money. However, if we consider that "waste" word is placed wrongly and if that be used as a "resource" then resource recovery from wastes is an emerging as thrust area of research and management because it offers environment and social sustainability potentials.

Many research groups are focusing their work on recovery of various resources such as energy, bio-products, nutrients (nitrogen and phosphorous), metals from wastewater generated by anthropogenic activities (Li and Yu 2011; Honse et al. 2012; Varjani and Upasani 2017; Mohan et al. 2018; Thakur et al. 2018; Varjani et al. 2019). Valuable pollutants are present in petroleum industrial wastewaters, which can be regarded as resources after recovery (Al-

Futaisi et al. 2007; Hu et al. 2013; Jasmine and Mukherji 2015). On one side efficient resource recovery and reuse can create sustainable livelihood whereas on the other side it supports green economy by reducing waste and improve environmental health and cost of recovery. Hence, there is a need to recycle and reuse the waste produced from activities of petroleum industry in an efficient manner. Feasible techniques to produce pollution less products create a new way for environmental and economic sustainability.

To optimize the exploitation of petroleum industry waste/wastewater and by-products there is a need to develop sustainable technologies. Focus of the research shall also be thrown on biorefinery concept for development of innovative bio-based industries because waste biorefineries may open up new market opportunities for bio-based products and achieve efficient resource utilization.

7) Conclusion

Petroleum industry wastewater can be treated by various physical, chemical and biological treatment processes. There are many hazardous components present in waste/wastewater released by the activities of petroleum industry. Successful remediation strategy should be tailored considering environment and human health. Bioremediation is one of the technologies which is gaining a global interest for cleanup of petroleum hydrocarbons. However, integration of various processes gives better results than individual process used for waste/wastewater treatment. The current practice of industrial wastewater treatment is focused to remove pollutants from wastewaters to meet the discharge standards. To resolve hazards associated with petroleum components, a suitable technology that treats waste and generates value addition would be a promising option. Study regarding the recovery of value-added products from waste/wastewater with special reference to different techniques, either separately or by integration, tailoring distinct features of processes are thrust area of research with respect to waste characteristics for production of bio-based non-toxic by-products. Waste biorefinery concept using latest developments in biotechnological and bioengineering options pertaining to recovery of resources from petroleum industry waste/wastewater shall also be explored.

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Table Legend

Table 1. Characteristics of oily wastewater/effluents and environmental standards for their discharge

Table 2. Petroleum Industry Wastewater Treatment Technologies

Table 1. Characteristics of oily wastewater/effluents and environmental standards for their discharge (Source: G.S.R 186(E), dated 18th March 2008)

Sr. No.	Parameter	Limiting value for concentration*
1	pH	6.0-8.5
2	Chemical oxygen demand	125.0
3	Biological oxygen demand 3 days, 27°C	15.0
4	Oil and Grease	5.0
5	Suspended Solids	20.0
6	Total Kjeldahl nitrogen	40.0
7	Sulphides	0.5
8	Phenols	0.35
9	Cyanide	0.20
10	Benzene	0.1
11	Benzo[a]pyrene	0.2
12	Metal contents	
	Hexavalent Chromium	0.1
	Total Chromium	2.0

Lead	0.1
Mercury	0.01
Nickel	1.0
Zinc	5.0
Copper	1.0
Vanadium	0.2

*Limiting value for concentration is in mg/L, except for pH

Table 2. Petroleum industry wastewater treatment technologies (Source: Dimoglo et al. 2004; Lin et al. 2001 2000; Vaiopoulou et al. 2005; Knight et al. 1999

Sr. No.	Name of the techniques used	Treatment details
1	Electrochemical technologies	To remove turbidity, COD, phenol, hydrocarbon and grease by using electro coagulation and electro flotation from petroleum wastewater
2	Ozonation and biological activated carbon (BAC)	To enhance biodegradation process of bio refractory and the growth of a biofilm have been noticed during laboratory scale pre-ozonation and biological activated carbon (BAC)
3	Anaerobic treatment	Anaerobic digestion method has been used for treatment of hydrocarbon pollutants
4	Aerobic sequencing batch reactors	Observer-based time-optimal control: Control strategy regulates and maintains the substrate concentration and feed ratio in the

		reactor. An extended Kalman filter has been used as a nonlinear observer in a petrochemical wastewater treatment
5	Autotrophic denitrification	To remove sulfide from petroleum industry waste water, new alternative treatment for removal of H_2S by combination of the biological method and existing stripping CO_2
7	Wetlands	Some large-scale project and pilot scale studies have been conducted for extraction and pumping stations of oil and gas for treatment using wetland for oil refineries

Figure Captions

Figure 1. Petroleum industry wastewater treatment

Figure 2. Treatment of wastewater of petroleum industry: Schematic diagram

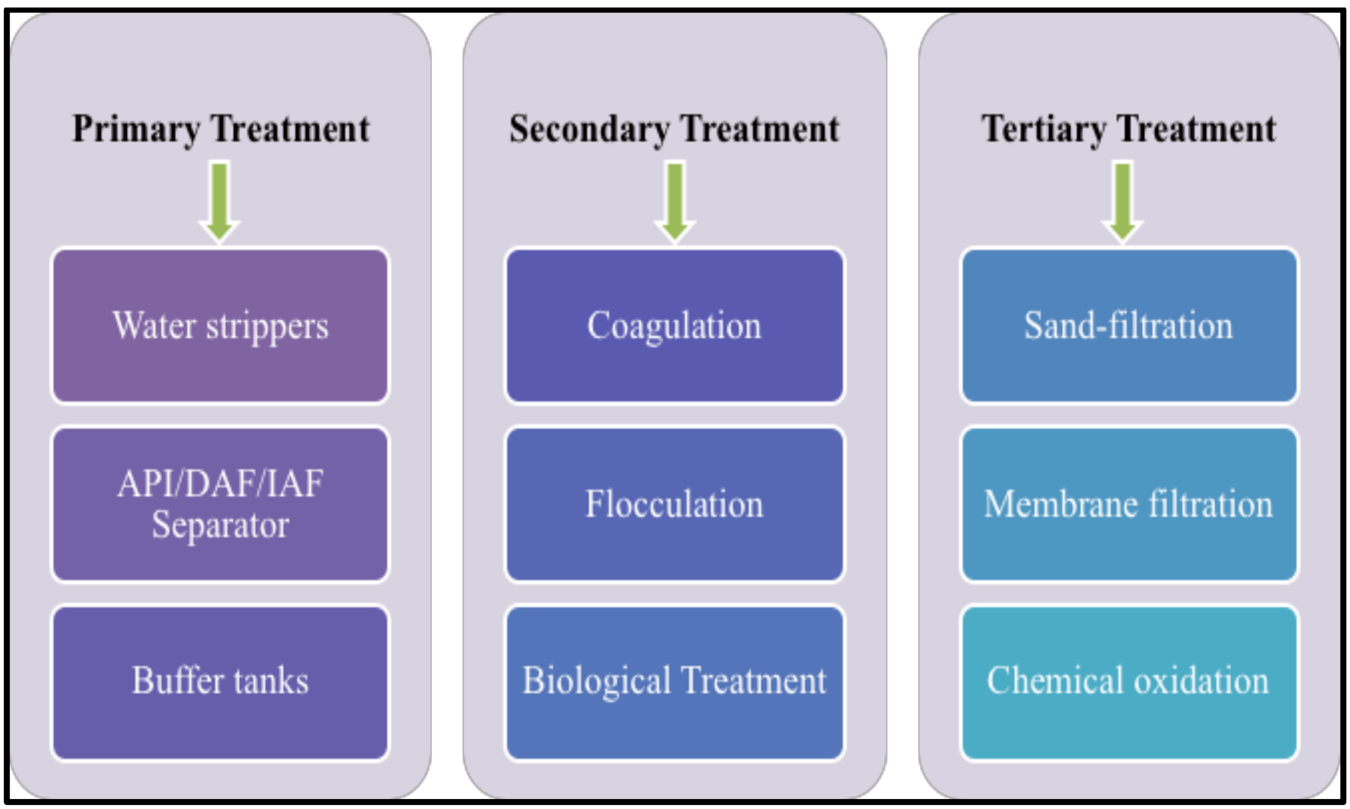

Figure 1. Petroleum industry wastewater treatment

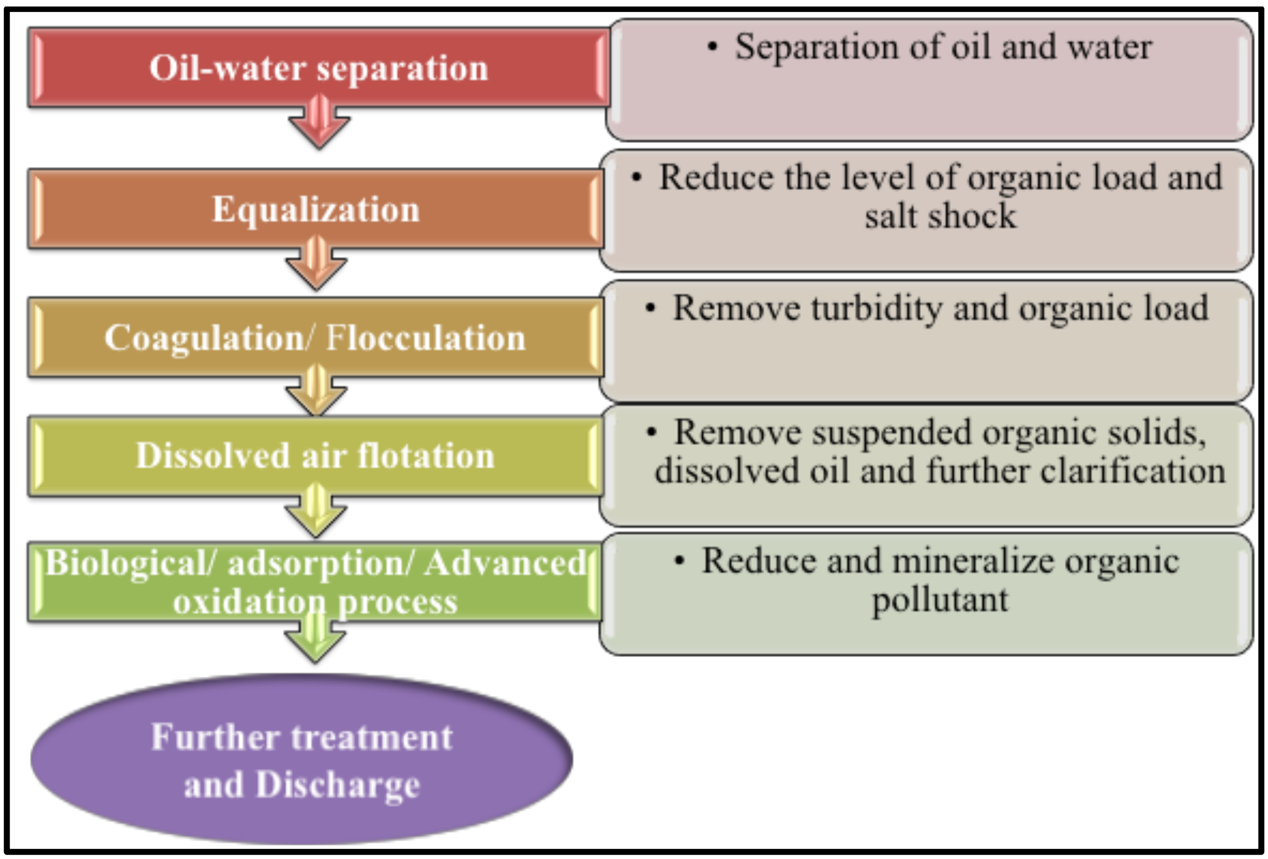

Figure 2. Treatment of wastewater of petroleum industry: Schematic diagram