

Review

A Review on the Treatment of Petroleum Refinery Wastewater Using Advanced Oxidation Processes

Wamda Faisal Elmobarak ¹, Bassim H. Hameed ¹, Fares Almomani ^{1,*}  and Ahmad Zuhairi Abdullah ²

¹ Department of Chemical Engineering, Qatar University, Doha P.O. Box 2713, Qatar; we1609816@student.qu.edu.qa (W.F.E.); b.hammadi@qu.edu.qa (B.H.H.)

² School of Chemical Engineering, Engineering Campus, University Sains Malaysia, Nibong Tebal 14300, Pulau Pinang, Malaysia; chzuhairi@usm.my

* Correspondence: falmomani@qu.edu.qa; Tel.: +974-4403-4140

Abstract: The petroleum industry is one of the most rapidly developing industries and is projected to grow faster in the coming years. The recent environmental activities and global requirements for cleaner methods are pushing the petroleum refining industries for the use of green techniques and industrial wastewater treatment. Petroleum industry wastewater contains a broad diversity of contaminants such as petroleum hydrocarbons, oil and grease, phenol, ammonia, sulfides, and other organic composites, etc. All of these compounds within discharged water from the petroleum industry exist in an extremely complicated form, which is unsafe for the environment. Conventional treatment systems treating refinery wastewater have shown major drawbacks including low efficiency, high capital and operating cost, and sensitivity to low biodegradability and toxicity. The advanced oxidation process (AOP) method is one of the methods applied for petroleum refinery wastewater treatment. The objective of this work is to review the current application of AOP technologies in the treatment of petroleum industry wastewater. The petroleum wastewater treatment using AOP methods includes Fenton and photo-Fenton, H₂O₂/UV, photocatalysis, ozonation, and biological processes. This review reports that the treatment efficiencies strongly depend on the chosen AOP type, the physical and chemical properties of target contaminants, and the operating conditions. It is reported that other mechanisms, as well as hydroxyl radical oxidation, might occur throughout the AOP treatment and donate to the decrease in target contaminants. Mainly, the recent advances in the AOP treatment of petroleum wastewater are discussed. Moreover, the review identifies scientific literature on knowledge gaps, and future research ways are provided to assess the effects of these technologies in the treatment of petroleum wastewater.

Keywords: petroleum wastewater refinery; advanced oxidation process; fenton; photo-Fenton; ozonation



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1. Introduction

Water is crucial for life since it is a fundamental necessity of all organisms. Fast industrial and economic growth have guided a rapid increase in population and development [1]. The world is observing growth and development in the industry, owing to the several industrial methods that are applied [2]. The industrial regions incessantly produce significant quantities of wastewater at high rates and generally dispose of the wastewater with no appropriate management and treatment [3,4]. Petroleum refineries and industries are crucial from an economic development perspective [5]. Petroleum refineries are meeting the difficulty of how to dispose this wastewater.

Wastewater produced by petroleum industries includes various kinds of organic and inorganic contaminants, for example sulfides, phenol, BTEX, hydrocarbons, heavy metals, etc. [6]. Huge amounts of harmful materials are produced through petroleum industry activities, for example through the oil production process, oil refinery, transportation, storage, etc., which are all unsafe for the environment and human health [6]. The treatment of wastewater produced from the petroleum industry includes different processes that

comprise physical, chemical, and biological methods [7]; however, the majority of these techniques are ideal to match certain treatment requirements for every application and generally do not recommend the treatment to separate the various groups of compounds [8].

The chemical oxidation method is one of the techniques used for wastewater treatment, and it can be classified into two types: conventional chemical treatments and advanced oxidation processes [9]. Advanced oxidation methods are highly efficient techniques needed for the treatment of several wastewater types, including the oil and gas industry, pharmaceutical industries wastewaters, etc. In previous years, several works have been carried out to examine the efficiency of advanced oxidation processes in the treatment of different wastewater types that contain recalcitrant and toxic pollutants [10]. AOPs are defined as the methods that depend on the production of hydroxyl free radicals, which have great electrochemical oxidant power and strong oxidizing potential [11,12]. The great oxidizing potential allows them to easily oxidize and degrade nearly all organic compounds to H₂O, carbon dioxide (CO₂), and inorganic ions, through dehydrogenation or hydroxylation. In general, AOPs are extensively employed in petroleum wastewater treatment for decreasing organic materials, the removal of certain contaminants, and sludge treatment [13]. The possibility of AOPs being used for wastewater treatment is improved by their capability to generate hydroxyl radical (OH•) via various methods.

The very common AOP method types studied for wastewater treatment involve heterogeneous photocatalytic oxidation [14], ozonation processes (O₃) [15], Fenton and photo-Fenton reaction [16,17], and electrochemical oxidation [18]. These methods are favorable in the treatment of wastewater containing different organic and inorganic pollutants. According to previous reviews, several combinations of various AOPs are more effective in comparison to a single oxidation process, owing to the production of additional radicals and excessive energy efficiency. Moreover, the utilization of heterogeneous and homogeneous catalysts and energy-dispersing elements considerably improves the degradation of the wastewater. Wang et al. [19] summarized hydrogen peroxide (H₂O₂), O₃, ultraviolet (UV)/H₂O₂ or O₃, O₃/H₂O₂, and photo-Fenton processes and their use in wastewater treatment. Furthermore, Chen et al. [20] and Ani et al. [21] reported a review on the basics of the heterogeneous photocatalytic degradation of organic pollutants and stated the benefits of utilizing titanium as an effective material for the removal of organic contaminants by applying advanced oxidation processes. Azizah and Widiyasa [22] reviewed the application of H₂O₂/UV and H₂O₂/UV/O₃ advanced oxidation process methods in synthetic residual fluid catalytic cracking (RFCC) wastewater to verify the suitable condition of AOPs to meet the phenol separation ranges. It is found that a high phenol degradation of 93.75% was achieved by using the H₂O₂/UV/O₃ AOP process with a H₂O₂ amount of 1000 ppm; this clarified that the H₂O₂/UV method requires extended time and greater H₂O₂ amounts for the removal of phenols from these wastewater types to levels below the target. Lofrano et al. [23] propose a generally updated review of AOPs in antibiotics removal considering physio-chemical and toxicity implications. They discussed the technical and economic problems, and the environmental compatibility of effluents.

An environmentally friendly preparation of TiO₂ nanoparticles supported by microwaves from titanium tetrachloride and water was carried out by de Oliveira et al. for the treatment of petroleum refinery effluent [24]. The highest rates of TOC separation were achieved below pH conditions of 10 and a catalyst dosage of 100 mg/L in 90 min of reaction time. The catalyst demonstrated steadiness for four cycles. The attained data revealed that the produced catalyst is a favorable material for the separation of remaining recalcitrant organic material from petroleum refinery effluent.

As far as the authors are aware, there are different studies on the treatment of petroleum refinery wastewater using the different types of an advanced oxidation process method as a tertiary treatment to disinfect and degrade the wastewater compounds and reuse the treated wastewater for different purposes. Therefore, the objective of the review is to present the different methods of the advanced oxidation process and its use in petroleum refinery wastewater treatment. The review reports that the treatment efficiencies depended

strongly on the selected AOP method type, whether the properties of target contaminants were physical or chemical, and the operating conditions. Moreover, the knowledge gaps and future research methods are also provided to assess the effects of these technologies in the treatment of petroleum wastewater.

2. Methodology

The review provides an overview of the findings from recent studies from 2017 to 2021 which have been applied to different types of AOPs to degrade and remove different compounds from petroleum refinery wastewater. Few articles published from 2012–2015 were also considered as they presented crucial data. The major source of these articles was google-scholar and Scopus databases; the keywords applied for the search were “advanced oxidation”, “petroleum”, “refinery”, “wastewater pollutants”, “Fenton”, “catalyst”, and “review”. The assembled data were introduced in Tables 1–5 and studied in four sections. Information on the characteristics of petroleum refinery wastewater and the environmental impact of petroleum refinery wastewater was presented in Tables 1 and 2, and discussed in Sections 3 and 4, respectively. The fundamentals of AOPs were discussed in Section 5, an overview of the process in petroleum refinery wastewater treatment was represented by Figure 1, and the classification of the AOPs was presented in Table 3. In this section (Section 5) four types of AOPs including H₂O₂/UV, Fenton, photo-Fenton, and ozonation AOPs and their applications in petroleum refinery wastewater treatment were discussed. The data relating to each process was presented in Tables 4 and 5, and Figures 2–6 were included to present crucial concepts related to each process (the reaction mechanism, the drawbacks of Fenton, and photo-Fenton processes, solar photoreactor setup, experimental setup of photoreactor, and detailed steps of the mechanism of organic degradation of wastewater contaminants through O₃/UV) [25,26]. In Section 6, the integrated AOPs were reviewed, and a diagram of the integrated system was presented in Figure 7. Finally, from the analysis of the assembled data, conclusions are presented and recommendations for additional research are provided.

3. Characteristics of Petroleum Refinery Wastewater

Petroleum refinery effluent has numerous amounts of complex substances present in it. They were present in the form of oil, gas, wax, grease, metals, minerals, hydrocarbons, etc. Few components present in very low amounts; such components are N, S, O, and a few other metals which have minor importance. Aromatic organic compounds containing solely carbon and hydrogen atoms are colorless, white, or pale-yellow solids. It has been proven that, when it enters the human body, our potential immune system will suppress it. This can cause carcinogenic and mutagenic damage [27].

The constituents present in the produced include oil and gases and different organic and inorganic matters that are complex in nature. The particles implicated are hydrocarbons, dissolved oil, dissolved salts [28], naturally formed radioactive elements, and heavy metals [29]. Furthermore, with gases such as hydrogen sulfides, carbon dioxide composition will be encompassed as dissolved gases. The generation of produced water is strictly controlled by different countries because the components present in it will increase the pollution rate [30]. The concentration of salt present in seawater is lower than compared to the concentration of salt present in the produced water. The percentage of salinity is enhanced due to the concentration of the additional component present in it. Those components are Ca, K, Mg, and dissolved Cl, etc. [31].

The previous survey [31] reveals that the electrical conductivity of the produced water lies in a range from 200 μ S/cm to 300 μ S/cm, while the TDS concentration range of produced water lies in a range from 120 g/L to 140 g/L. Some of the major compositions are sodium with a range of (42,720 \pm 2093 mg/L), calcium ions (4247 \pm 752 mg/L), magnesium ions (727 \pm 54 mg/L), potassium ion (805 \pm 230 mg/L), strontium ions (257 \pm 20 mg/L), chlorine ions (65,800 \pm 1600 mg/L), sulphate (1010 \pm 9 mg/L), bromide ion (591 \pm 16 mg/L) and silicon dioxide (32 \pm 2 mg/L) [27]. The concentration of ammonia present

in the PW has a range of $(655 \pm 77 \text{ mg/L})$ [31]. The value of pH, as well as alkalinity, are 7.30 ± 0.21 and $2345 \pm 329 \text{ mg/L}$, respectively, such as CaCO_3 [32]. Table 1 shows the composition present in the petroleum refinery wastewater [33]. The composition of nitrogen in the produced water coming from the oil platform is 50 v/v%. The summation of all nitrogen compounds in the water is termed total nitrogen; it contains ammonia nitrogen ($\text{NH}_3\text{-N}$), nitrite nitrogen ($\text{NO}_2\text{-N}$), nitrate nitrogen ($\text{NO}_3\text{-N}$), and organically bonded nitrogen [34].

Table 1. The compositions present in the petroleum refinery wastewater.

Composition of Each Element	Composition in Petroleum Wastewater	Limitations	Metal Compositions	Components in Petroleum Wastewater	Limitations
pH	4.3–10	6.5–8.5	Ca	18–132,687	100
Density	1014–1140	-	Na	316–134,652	-
TOC	3.4–5960	-	K	8.6–14,649	-
COD	1200	≤ 50	Mg	4–18,145	100
BOD	-	≤ 30	Fe	<0.1–100	0.3
TSS	1.2–21,820	30	Al	310–410	≤ 0.2
TDS	1×10^3 – 4×10^5	1200	B	5.00–95	1
DO	8.2	<4.0	Ba	0–22400	1
TPH	2–565	-	Cd	<0.005–0.2	0.005
BTEX	0.39–35	-	Cr	0.02–1.1	≤ 0.1
base and neutrals	<140	-	Cu	<0.002–1.5	1.3
Cl	80–310,561	1400–190,000	Li	3–50.00	-
Br	0–12,000	150–1149	Mn	<0.004–175	0.05
I	0–500	-	Ni	-	0.3
HCO_3	1.9–7355	-	Pb	0.002–8.8	0.015
CO_3	0–800	-	Sr	0.02–1000	-
SO_4	0.5–7851	<0.1–47	Ti	<0.01–0.7	-
PO_4	0–0.10	-	Zn	0.01–35	7.4
SO_3	~10	-	As	<0.005–0.3	0.02
NO_3	0–3.5	-	Hg	<0.001–0.002	0.005
NO_2	0.05	-	Ag	<0.001–0.15	0.1
$\text{NH}_3\text{-N}$	10–300	-	Be	<0.001–0.004	-
Total Polar	9.7–600	-	NORM (pCi/L)	-	-
Higher acids	<1–63	-	Total Ra	0.054–32,400	5
Phenols	till 23	-	U	0.008–2.7	-
VFA	2–4900	-	Th	0.008–0.027	15
Oil & Grease	6.9–210	2.3–60	Pb	1.35–5130	-
m-xylene	0.01–54	-	Po	0.005–0.17	-
MBAS	0.01–54	-	-	-	-
HEM	0.6–2000	0.02	-	-	-
Alkalinity	6.1–200	-	-	-	-

The configuration of these substances and their tenacious character affects the rate of chemical oxygen demand and the level of toxicity. The wastewater encompasses a high level of COD. The presence of these toxic oils considerably disperses the chemical oxidation demand [32]. If the produced water contains a high content of BOD value, it is due to the large content of organic materials present in the drilling fluids. Before discharging a high value of BOD content water into the water bodies, oxidation of the water must be maintained so that only it can resist the wastewater with a larger BOD content [29]. The PW containing the range of the TOC is (0–1500 mg/L). The naturally formed water has a TOC concentration that lies in the range of <0.01–11,000 mg/L [35]. The World Health

Organization proposes that a fringe of the permissible volume of benzene and toluene in drinking water are 10 and 700 mg/L, respectively, while the permissible proportion of ethylbenzene and xylene is 300 mg/L and 500 mg/L, respectively [27].

The characteristic of the petroleum refinery wastewater fluctuates according to their base. The constitution of the wastewater depends on the final product induced, the undertaking process, and the type of crude oil that is utilized. The produced water is assorted with aromatic and aliphatic hydrocarbons, MTBE, PAHs, phenols, naphthenic acid, sulfides, metal products as well as benzene, toluene, ethylbenzene and xylene (BTEX) that usually possess a high toxicity. In addition, xylene has three more topical isomers named o-xylene, m-xylene, and p-xylene [36].

4. Environmental Impact of Petroleum Refinery Wastewater

Enormous amounts of contaminants were released by petroleum products in the environment [37]. As known, petroleum wastewater is the main of three waste types (wastewater, waste gas, and industrial residue) whose harmful impacts cannot be misjudged [38]. Wastewater discharged by petroleum industries include significant amounts of heavy metals, petroleum hydrocarbons [39], polyaromatic hydrocarbons [40], benzene, toluene, ethylbenzene, xylene (BTEX), phenolic compounds, and other contaminated materials [41]. Some of the major petroleum wastewater contaminants and their environmental impacts are presented in Table 2. Owing to the incompetence of treatment methods, petroleum refinery wastewater is dangerous to the environment and other life systems which could become mutagenic and toxic to human beings [42]. This wastewater can influence various elements of the environment, for example clean water, human health, the different sources of groundwater, air, marine life, crop manufacturing, etc. [43]. An increase in contaminants in the water bodies raises significant concerns, either long-term or short-term, for the ecology and living organisms, which can be severe and chronic [44]. Different studies revealed that petroleum refinery wastewater composites have the ability to biologically build up, which is the main reason to create a method for the separation of pollutants from water before the release [39]. The following problems were created when a huge quantity of petroleum oily wastewater was arbitrarily released into water bodies [7].

Regarding aquatic life where the wastewater was released into the river and oceans, oil and other contaminants create a film on the water surface that lead to a block in the air and sunlight, thus causing a shortage of oxygen for the aquatic organisms and affecting the plants and animals' growth [45]. Furthermore, owing to the existence of heavy oil on the surface of seawater, serious problems were observed, including slow growth of the head of aquatic organisms, unusual neural growth in the embryo of the aquatic organisms, and other issues [46]. Benzene, toluene, ethylbenzene, and xylene (BTEX) are some of the main pollutants that harm aquatic organisms, the nervous system, genotoxicity, and the respiratory system [46], due to their highly carcinogenic nature as distinguished by The U.S. Environmental Protection Agency (USEPA) [42]. Moreover, petroleum refinery wastewater includes a solvable aromatic hydrocarbon, many radioactive substances, and heavy metals which can generate negative impacts on some organisms accountable for water purification and biological treatment [36]. In the case of direct release, pollutants in the wastewater are transported by the food chain through the infected foods, and, hence, human health will be critically affected [47]. Aquatic contamination will also occur, owing to the spread of toxic components between the food chain via the marine ecosystem from the influenced marine environment [48]. Some studies reported that the consumption of the contaminated oil with marine food could produce many toxic consequences on human health, such as skin tumors due to the existence of the contaminated hydrocarbons in the polluted wastewater [49]. Another study presented the effect of wastewater pollutants during the separation processes of oil throughout the marine water [40]. It was found that the co-workers that engaged in certain processes developed a urinary infection due to the presence of a high concentration of volatile organic composites and polycyclic aromatic hydrocarbons which can be absorbed by their body. Furthermore, it was found that

contaminated oil in the wastewater represents a toxic material to certain microorganisms that is accountable for the biological treatment of wastewater [50].

Table 2. Some of the main wastewater contaminants mentioned in the work and their environmental impacts.

Contaminant	Environmental Impact	Ref
Phenol	Have harmful effects on the muscles, causing moving problems, pain to the gastrointestinal system, and even death.	Yang et al. (2020)
Methyl-butyl ether	In an aquatic environment, it quickly causes the reduction in dissolved oxygen. Regarding human health, it causes kidney and blood cancers and affects the nerve. The existence in the aquatic environment affects the stability and health of the organisms, leading to a low regenerative rate, and affecting the behavior of the organisms. It also reduces the growth of plants and animals, and, with long time exposure, it can lead to their death.	Gallo et al. (2020)
Benzene	Have harmful impacts on the different types of microorganism (aquatic, soil, etc).	Rabani et al. (2020)
Toluene	Exposure for long periods affects the kidney which can lead to pain in the inner ear. Affects the kidney, nervous system, and the dysfunctions of the liver. In addition, various negative impacts on the neurological system have been stated.	Poyraz et al. (2020)
Ethylbenzene		Wollin et al. (2020)
Xylene		Egendorf et al. (2020)

Phenols are other toxic compounds that generally appear in the wastewater produced from petroleum refinery and can impact the enzymatic processes of marine microorganisms even at small phenol concentrations. As the degradation of the phenolic composite takes a long time, they will gather in the aquatic organisms' tissues and consequently produce biomagnification. The phenolic compounds have been identified to be genetically, physiologically, and haematologically toxic [51]. Thus, the phenolic compounds have been classified by USEPA as a significant contaminant [52]. The phytotoxicity of phenols was experimentally verified on *EU cyclops agilis* by Kottuparambil et al. [53]. The results showed the high toxicity of phenol compounds even in the presence of a small dose of phenols and how this negatively impacts human life [54].

Polycyclic aromatic hydrocarbon (PAHs) compounds are other toxic contaminants that existed in the petroleum industry wastewater and have toxic effects on aquatic organisms. The study reported by Badibostan et al. [55] showed that the presence of specific polycyclic aromatic hydrocarbons, for example chrysene (CHR) compounds and benzo[b]fluoranthene (BbF) with high concentrations, causes serious problems including drinking, inhalation, etc. When its overall concentration exceeded the concentrations of the standards defined by USEPA for toxic chemicals, it negatively impacted human life and health.

Another concern is that, when petroleum oily wastewater used for irrigation, these compounds can have harmful impacts on the soil and make it stronger [56]. This change on soil texture inhibits the breathing of plants and organisms as the soil microorganisms have a high sensitivity to MTBE compared to the microorganisms present in the water. In addition, oily wastewater could have a harmful effect on environmental processes such as nutrient cycles and biodegradation [56]. Consequently, the microorganism's decline rate will decrease, and then the plants and animal growth will be influenced [57].

5. Advanced Oxidation Process (AOPs) for Petroleum Refinery Wastewater

5.1. Fundamentals/Chemistry of the Advanced Oxidation Process

Advanced oxidation processes (AOPs) are the methods defined through the creation of hydroxyl radicals; the core purpose of the advanced oxidation process is to produce highly reactive free radicals which are extremely sensitive, and to apply non-selective materials to reduce contaminated organic composites that exist in a medium for example wastewater, soil, marine mediums, etc. [58].

Hydroxyl radicals are efficient in eliminating organic chemicals since they are reactive electrophiles [59]. They are powerful oxidizing agents to demolish compounds that cannot be oxidized by the standard oxidant. Hydroxyl radicals have a more rapid oxidation rate compared to KMnO_4 or H_2O_2 [60]. The produced hydroxyl radicals can remove the organic chemicals through different ways such as radical addition, hydrogen extraction, or electron transmission [61]. The creation of hydroxyl radicals is generally enhanced by merging O_3 , H_2O_2 , TiO_2 , UV radiation, ultrasound, and electron-beam irradiation [62].

The hydroxyl radical has great oxidation potential ($E_0 = 2.8 \text{ V}$) [63] and is competent to react with almost all types of organic composites, leading to full mineralization of these composites which form from carbon dioxide (CO_2), inorganic salts, and water, or their transformation into fewer destructive substances [64]. The removal of contaminants and the inhibition of the production of toxic compounds are some of the major advantages of AOPs since traditional techniques of water treatment. For example filtration, adsorption, and flotation are not effective in completely separating the organic pollutants (non-destructive physical separation procedures), which only eliminate the pollutants, transporting them to other stages, and thus producing intense residues [65]. An overview of the AOP in petroleum wastewater treatment is presented in Figure 1.

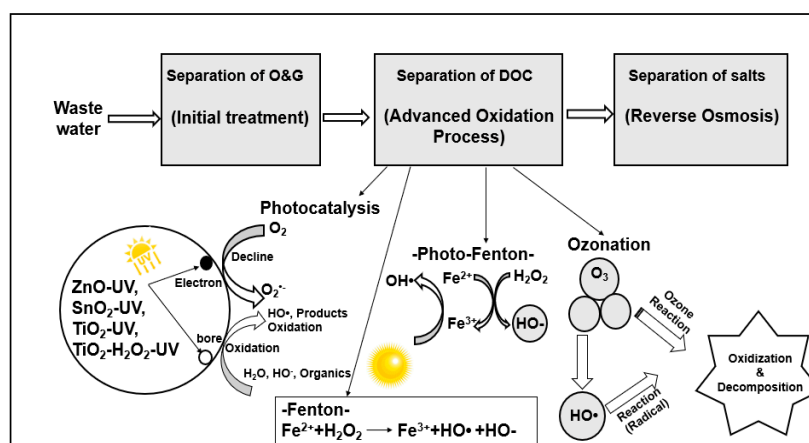
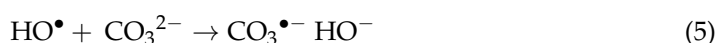
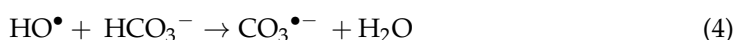


Figure 1. An overview for the AOP in petroleum refinery wastewater treatment.

The main reaction produce hydroxyl radicals ($\text{HO}\bullet$) that attack organic composites in electrophilic position leading to degradation. In addition $\text{HO}\bullet$ reaction with organic composites (aromatic or unsaturated) that include a π bond result in the formation of the organic radicals (Equation (1)) [66]. This includes hydrogen removal through the reaction of the $\text{HO}\bullet$ radicals with a saturated aliphatic compound (Equation (2)) and electron transmission by decreasing the $\text{HO}\bullet$ radical into a hydroxyl anion through an organic

substrate (Equation (3)) [67]. The attack of the HO• radical on organic substrates could be affected by the existence of different chemical types in water (which are created in the mineralization method), for example, bicarbonate and carbonate ions [68]. These ions can react with the HO• radicals (Equations (4) and (5)), and, by consequently vying with the organic substrates via the HO• radicals. HO•, radicals can be created from numerous AOPs, allowing the use of a superior method for every treatment condition.



Following Flouret and his colleagues (2018), the AOP can be classed as homogeneous and heterogeneous [69]. They introduced this categorization in terms of the light type that is utilized in the method. Table 3 shows the categorization of AOPs consistent with these studies. The AOP are costly methods due to the high cost of chemicals applied (e.g., H₂O₂ and O₃), also due of electric energy for UV radiation. Hence, they are studied in other treatments of wastewater that cannot be organically handled. An additional substantial part is the weight of contaminants that appear in the wastewater, commonly identified according to the existing amounts of COD. The application of these methods is revealed solely for wastewater that contains COD under 5 g/L, since a greater index of COD inhibiting the treatment would need an extremely higher utilization of chemicals [70]. For high organic wastewaters, pretreatment processes, such as dilution, coagulation, and flocculation, are needed to lessen the initial concentrations [71]. Additionally, AOPs can be utilized for treating the polluted waters containing very small organic contents (in ppb) which include dispersed organic composites that are hard to eliminate. One method of decreasing the AOPs' costs is to utilize them as initial treatment processes to decrease harmfulness, supported by biological treatment. This alternative has demonstrated to be very exciting from an economic perspective and has been examined by numerous studies [72,73].

Table 3. Types and categorization of advanced oxidation methods (AOPs).

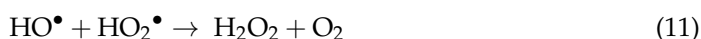
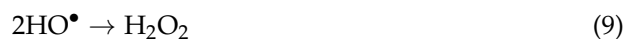
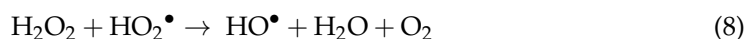
Non-photochemical-Homogeneous Methods	Photochemical-Homogeneous Methods	Non-Photochemical-Heterogeneous Processes	Photochemical-Heterogeneous Processes
<ul style="list-style-type: none"> Ozonation in alkaline media (O₃/HO⁻) Ozonation with hydrogen peroxide (O₃/H₂O₂) Fenton (Fe²⁺ or Fe³⁺/H₂O₂) Electro-oxidation Electrohydraulic discharge-ultrasound Wet air oxidation (WAO) Supercritical water oxidation (SCWO) 	<ul style="list-style-type: none"> Photolysis of water in vacuum ultraviolet (VUV) UV/H₂O₂tyg,v = UV/O₃ UV/O₃/H₂O₂ Photo-Fenton Fe²⁺ or Fe³⁺/H₂O₂/UV 	<ul style="list-style-type: none"> Catalytic wet air oxidation (CWAO) 	<ul style="list-style-type: none"> Heterogeneous photocatalysis: /UV, SnO₂/UV, TiO₂/UV, TiO₂/H₂O₂/UV

5.2. H₂O₂/UV Advanced Oxidation Process and Their Application in Petroleum Refinery Wastewater

Hydrogen peroxide (H₂O₂) is a powerful oxidant among other oxidant types with a standard reduction potential = 1.77 V that is used to decrease the small levels of contaminants that exist in wastewaters [74]. However, the specific utilization of H₂O₂ is not effective in treating more compounds and mutinous substances with a low reaction rate. The application of H₂O₂ is more successful when it performs in combination with other components or sources of energy that are competent of separating it to produce HO• radicals, which can behave as oxidizing factors [75]. Through UV radiation in wavelengths >300 nm, hydrogen peroxide (H₂O₂) can decompose and create HO•, as demonstrated in Equation (6).



H₂O₂ can also react with HO• and created transitional products in line with the reaction, the method is explained easily by Equations (7)–(11) [76].



The damage on organic composites occurs when HO• and hydroperoxyl (HO₂•) radicals are created. Nevertheless, the hydroperoxyl radicals have a smaller reduction potential with a value of 1.7 V compared to hydroxyl radicals with a value of 2.8 V; as a result, their production is not attractive to the method. Numerous researchers have indicated that the increase in the first amount of H₂O₂ enhances the pollutants degradation rate up to the highest value, after which, when they achieve very high H₂O₂ levels, they start to decrease. Khan and coworkers (2020) ascribe this reduction in the H₂O₂/UV process outputs to hydroxyl radicals reacting with additional H₂O₂ (Equation (7)), rather than reacting by the organic substrates, resulting in the creation of the HO₂•. The H₂O₂ photolysis is generally achieved using different mercury vapor lights (low or medium pressure) with great intensity, which decreases the required quantity of H₂O₂. Almost 50% of the energy expended is wasted in heat form or in emissions of wavelengths >185 nm, which are noticed by the jacket made of quartz [77].

A cheaper and widely used alternative is the germicide lamp; though, the efficiency is smaller as it radiates in a range from 210 to 240 nm, and H₂O₂ absorption achieves a peak of 220 nm [78]. The application of the H₂O₂/UV process has different advantages that can be ascribed to different facts, for example: the H₂O₂ reagent is completely solvable in water, there is no restriction of mass transfer, it is an active supply of HO•, and there is no necessity for a removal method after the treatment [79]. The operating pH should be small (pH < 4) to void the impact of separating radical types, mainly ionic types such as bicarbonate and carbonate ions, which results in improving the degradation rate.

The applications of H₂O₂ was evaluated in the existence and the nonappearance of UV radiation, and in the degradation of petroleum refinery wastewater which is pretreated using different separation processes such as coagulation and flotation, studying total petroleum hydrocarbons (TPH), 1,2-dichloroethane (DCE), dichloromethane (DCM), and MTBE [80]. It was remarked that UV emission did not substantially impact the compound degradation, except for DCM which was highly stable compared to others. Overall, 83% was eliminated with 11.76 nM of H₂O₂ in the existence of UV radiation. The studies showed sluggish degradation of the TPH, which separated by 69% in the first 8 days of testing. They found that the degradation of MTBE was identical to that of DCM, and that total degradation occurred after 24 h. The application of the H₂O₂/UV process for

the purification of oily wastewater from a lubricant production unit was analyzed by Gilpavas et al. (2019). Chromatographic characterizations that were carried out in the study indicated that most of the composites detected in the wastewater were separated with the ending of the treatment, utilizing great amounts of H_2O_2 . However, a decrease in just 40% of preliminary wastewater COD was achieved, which is equivalent to around 9000 mg/L. This was assigned to the creation of organic acids, generated because of the decomposition of the organic compounds that exist in the wastewater, which are more resistant to photo-degradation with H_2O_2 . The influence of pH on the process was also evaluated and it was found that, in an acidic condition of pH 3.5, there was a greater decrease in the wastewater COD, in comparison to basic and neutral pH conditions [81]. The degradation of MTBE was investigated by Neisi and coworkers (2018), and the gasoline was evaluated as one of the highly popular pollutants in polluted underground waters. The H_2O_2 /UV process was evaluated at certain conditions, and the removal percentage of MTBE was found to be more than 98%. A rise in the removal percentage was obtained with an increase in the concentration of H_2O_2 , after which the percentage began to drop [82]. This result was assigned to the competitive reactions of H_2O_2 and $HO\bullet$, presented in Equations (7)–(11). A comparison between some studies applying H_2O_2 advanced oxidation-based process in the treatment of different types of wastewater was illustrated in Table 4.

Table 4. Comparison between recent studies using H_2O_2 advanced oxidation-based process in wastewater treatment.

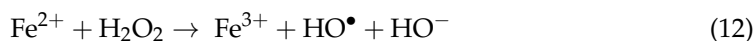
Process	Type of Wastewater (WW)	Conditions	Efficiency	Comments and Details	Ref
TiO_2 /UV	Model: TPH Benzene (Toluene Phenol) Naphthalene	$[TiO_2]$ 100 mg/L pH 6.5 for 1 h	COD reduction from 970 mg/L to 65 mg/L = 93% removal at 90 min, 30 °C and pH 3 using 100 mg/L	Higher degradations rates for with TiO_2 /UV = 92%, 98.8%, 91.5%, and 93% for B, T, P and N, respectively.	Ihtisham et al. 2020
H_2O_2 /UV	Model [Phenol] 100 mg/L	(H_2O_2) 3060 mg/L pH 9	99% phenol removal in 60 min	H_2O_2 /UV < O_3 /UV 0.081 min^{-1} < 0.0881 min^{-1}	Alrousan et al. 2020
H_2O_2 /UV	Oil COD 9000 ± 500 mg/L	(H_2O_2) 3330 mg/L pH 12	40% COD removal in 150 min	Negative impact on COD removal at basic pH	GilPavas et al. 2019
H_2O_2 /UV	Model: [nonylphenol] 4.41 mg/L	(H_2O_2) 1.7–17 mg/L pH 11	To achieve 90% of degradation [H_2O_2] 1.7 mg/L in 20 min (H_2O_2) 3.4 mg/L in 16 min (H_2O_2) 8.5 mg/L in 12 min	At pH 7 the time needed was 3 times higher in all concentrations studied	Kaur et al. 2020
H_2O_2 H_2O_2 /UV	Linear Alkyl Benzene COD 300–350 mg/L	(H_2O_2) 340 mg/L pH 9	H_2O_2 : 27% COD removal in 180 min H_2O_2 /UV: 36% COD removal in 180 min	H_2O_2 (27%) < O_3 /UV, H_2O_2 /UV (36%) < O_3 , (37%) < H_2O_2 / O_3 (39%) < H_2O_2 / O_3 /UV (42%)	Fernandes et al. 2019
H_2O_2 /UV	Model: MTBE 25 mg/L	(H_2O_2) 34 mg/L pH 12	98% MTBE removal in 60 min	H_2O_2 /UV (0.126 min^{-1})	Neisi et al. 2018

Table 4 shows a comparison between recent studies for wastewater treatment using H_2O_2 advanced oxidation-based processes at different conditions. The studies showed the high efficiency of the H_2O_2 -based advanced oxidation process in the degradation and removal of different wastewater compounds, such as COD, TOC, MTBE, phenol, etc., from different wastewater types. The efficiency was found to be more than 90% for each

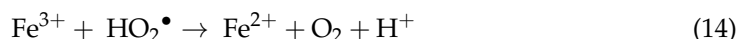
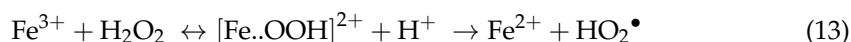
wastewater that either utilized the hydrogen peroxide H_2O_2 alone or added in the existence of ultraviolet (UV) light.

5.3. Fenton and Photo-Fenton Advanced Oxidation Processes and Their Application in Petroleum Refinery Wastewater

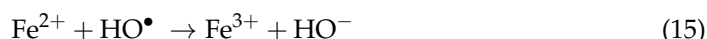
Fenton's chemical is a mixture of H_2O_2 and ferric ions found by Henry John Horstman Fenton (1854–1929). In 1894, he stated that this mixture had great oxidizing potency in an acid medium [83]. However, the use of Fenton's chemical in oxidation procedures to remove contaminated organic composites was reported by different researchers [84,85]. There is significant disagreement about the mechanism of reaction, including Fenton's reaction. The standard reaction of the Fenton process is taken by Haber and Weiss et al. (1934), as mentioned by Zouanti et al. [86], which contains an aqueous integration of hydrogen peroxide (H_2O_2) and ferrous ions (Fe^{2+}) in an acidic medium, leading to H_2O_2 breaking into a hydroxyl ion and a hydroxyl radical, and the oxidation from Fe^{2+} to Fe^{3+} , as characterized by Equation (12) [87].



The Fe^{2+} and Fe^{3+} types exhibited in an easy manner coincide with the $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^+$ aqueous compounds, which, combined with H_2O_2 , create the $[\text{Fe}(\text{OH})(\text{H}_2\text{O}_2)(\text{H}_2\text{O})_4]^+$ and $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^{2+}$ composites, respectively, as stated by [88]. The Fe^{3+} created in Equation (12) are able to react with the H_2O_2 that exists in the medium, and can be decreased to Fe^{2+} once more by producing the hydroperoxyl radical, according to Equation (13). This reaction happens more sluggishly than reaction 12, as described by Kim et al. [89]. The Fe^{3+} ions also react with the HO_2^\bullet and are decreased to Fe^{2+} , as indicated with Equation (14).

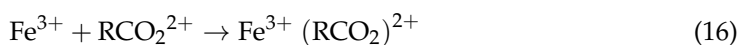


The preliminary rate of degradation employing Fe^{+3} is significantly smaller than that utilizing Fe^{+2} , as presented by Yuan et al. [90]. The standard pH in Fenton's reaction observed in several reports is 3 [91,92] and, consequently, can be considered the suitable operational pH. At an extremely low pH (<2.5), the creation of compounds, for example $[\text{Fe}(\text{H}_2\text{O})_6]^{2+}$, occurs. These react more sluggishly with H_2O_2 than $[\text{Fe}(\text{OH})(\text{H}_2\text{O})_5]^+$, generating a smaller volume of hydroxyl radicals and consequently reducing the efficiency of the system [93]. In the case of basic pH conditions, the reaction between the iron and hydroxide ions (HO^-) produced the iron hydroxide ($\text{Fe}(\text{OH})_2$ or $\text{Fe}(\text{OH})_3$) which does not react with H_2O_2 , thus prohibiting the Fenton reaction. Hence, a change in pH is necessary for wastewater treatment before the addition of Fenton reagents. Previously, it is crucial to determine the ideal correlation between H_2O_2 ions and ferrous ions. Once the ferrous ions were applied with higher concentrations more than hydrogen peroxide ions, the HO^\bullet created by Equation (12) can react with additional ferrous ions following Equation (15), and then reduce the raid of HO^\bullet on organic reacted material [94].

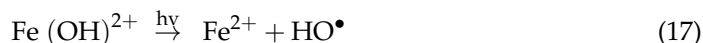


Wang et al. (2019) noticed that, while the ratio of $[\text{Fe}^{2+}]/[\text{H}_2\text{O}_2]$ is 1:2 and has a greater degradation rate, it is generally suggested to utilize a ratio of 1:5, which generates identical findings and needs fewer reagents. In general, only the reaction of the Fenton process is not able to depredate organic composites and mineralize them effectively. The reaction of the Fenton method is efficient up to the instant where each Fe^{2+} that exists in the reaction is reacted to Fe^{3+} , hence disrupting the production of hydroxyl radicals and subsequently degrading the organic compound [95]. Wang et al. [89] mentioned that one of the parameters that have a great effect in disturbing the Fenton's reaction under normal

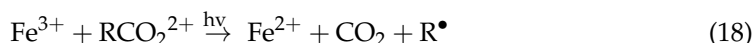
conditions with the degradation products Fe^{3+} ions can produce steady organic composites, specifically the organic acids existing in the reaction [91], as revealed by Equation (16).



While Fenton's reaction has been extensively reviewed and has presented great results in the wastewater treatment process, the reaction of Fenton was identified as a strong means to reduce organic composites when UV radiation is applied to the system. This was accomplished in the initial 1990s when used for treating wastewaters that include polluted carbon-based contaminants. It has acted as an initial step for different researchers associated with the use of the photo-Fenton method [96,97]. It has observed that UV irradiation greatly increases the organic contaminants degradation rate from Fenton's chemical, which has the feature of being susceptible to UV-Vis emission for wavelengths over 300 nm. Below these situations, the Fe^{3+} compounds photolysis allows the existence of Fenton's reaction and the Fe^{2+} to be reused many times if H_2O_2 is presented. Clarizia et al. [98] stated that, in a pH range from 2.5 to 5, the ferric compound $\text{Fe}(\text{OH})^{2+}$ is the main type in the photo-Fenton method, a shortened form of demonstrating the aqueous composite $\text{Fe}(\text{OH})(\text{H}_2\text{O}_5)^{2+}$. The photolysis of this compound at wavelengths less than 410 nm is found to be the major $\text{HO}\cdot$ radical source (Equation (17)) [98]. Additional photoreactive classes also exist in the medium of the reaction, for example $\text{Fe}_2(\text{OH})_2^{4+}$ and $\text{Fe}(\text{OH})_2^+$, which could lead to the creation of hydroxyl radicals [99].



The regeneration of Fe^{2+} can also happen with the produced compounds photolysis (Equation (16)) from Fe^{3+} and the organic compounds created in the degradation method, as demonstrated by Equation (18) [89].



Hence, the ferrous ions reformed by Equations (17) and (18) can react once again with the H_2O_2 in solution (Equation (12)) to produce more hydroxyl radicals, forming a photocatalytic cycle in the $\text{Fe}^{2+}/\text{Fe}^{3+}$ system. The utilization of radiation considerably lowers the essential number of ferrous ions, in comparison to the dim Fenton reaction (lack of light). The number of ferrous ions applied in the Fenton's reaction must be reduced to attain good efficiency, because adding high concentrations of ferrous ions rises the opacity of the solution, prohibiting the radiation diffusion and the Fe^{2+} production when oxidized to Fe^{3+} . As a result, the rate of degradation is reduced [100]. It is also found that the elevated intensity of light dispersion and the strong connection between the contaminant and the oxidizing factor act as benefits of the photo-Fenton method because it is a consistent method [101,102]. Figure 2 shows the reaction mechanism for the photo-Fenton process.

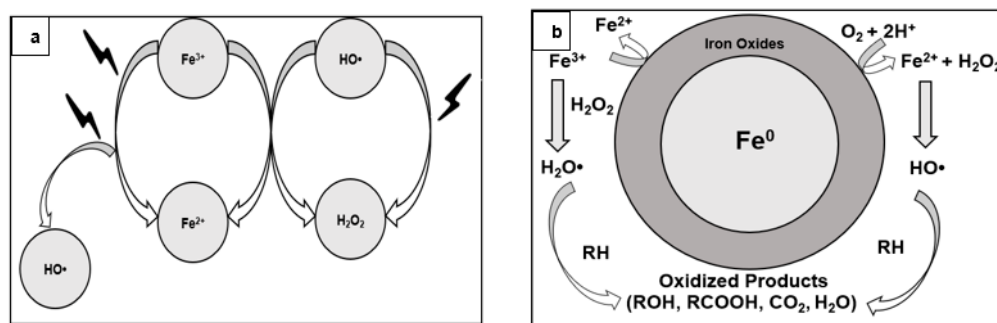


Figure 2. (a) Photo-Fenton, (b) Fenton process reactions mechanism.

The drawbacks of the Fenton and photo-Fenton processes are related to need for pH values which are too small (usually less than 2) and the need to eliminate the iron after reaction [103]. The solution pH in Fenton and photo-Fenton processes should be shifted to protect the catalyst stability, as at pH 6 iron hydroxide is normally generated. For various materials, the proper pH for the Fenton reaction is between 2.8 and 3, and the ideal ratio between catalyst and peroxide is generally 1:5 wt/wt. At an operational pH of >3 (acidic pH), the decomposition rate declines as a result of the reduction in free iron types in the solution, possibly owing to the creation of Fe(II) complexes with the barrier hindering the production of free radicals. At pH greater than 3, Fe^{3+} begins precipitating as ferric oxyhydroxides and breaks down the H_2O_2 into O_2 and H_2O [104], consequently decreasing the production of ferrous ions. Moreover, the $\bullet\text{OH}$ radical oxidation potential is known to reduce with a rise in pH [105]. Nevertheless, at low pH (pH = 2.5), the creation of $\text{Fe(II) (H}_2\text{O)}^{2+}$ happens, which reacts more slowly with hydrogen peroxide, generating a reduced volume of reactive hydroxyl radicals and consequently decreasing the degradation efficiency [75]. The main drawbacks for Fenton and photo-Fenton processes are summarized in Figure 3.

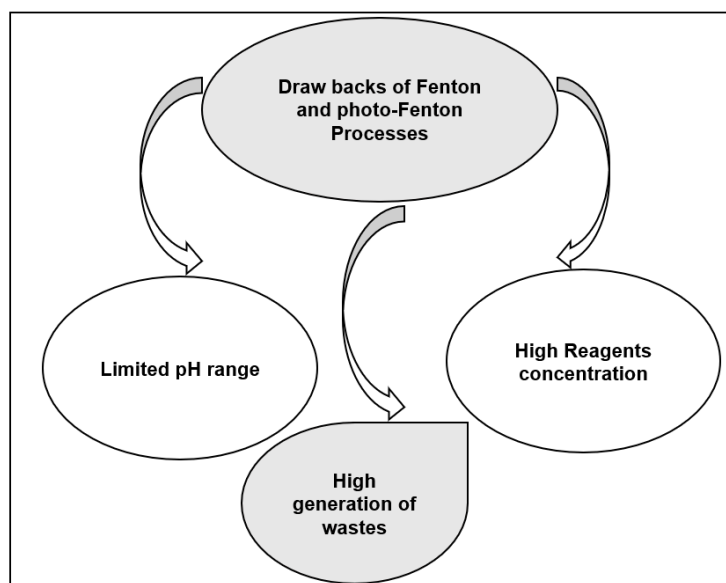


Figure 3. The major drawbacks for Fenton and photo-Fenton processes.

As stated by Rajala et al., [106] iron separation could not be required if it is applied with amounts under the removal range determined by regulation. It is crucial to have the previous familiarity of the original wastewater's physical and chemical features (untreated wastewater). Using this method, since some materials or inorganic ions for example Cl^- , $(\text{SO}_4)^{2-}$, $[(\text{H}_2\text{PO}_4)^- / (\text{HPO}_4)^{2-}]$ exist in the wastewater or are added as reagents such as $(\text{FeSO}_4, \text{FeCl}_3, \text{HCl}, \text{H}_2\text{SO}_4)$, may negatively affect the Fenton and photo-Fenton system reaction mechanism, thus hindering the degradation method [107]. The main reason for this is the scavenging effect of these ion (Fe^{2+} or Fe^{3+} or any other inorganic ions) to $\text{OH}\bullet$ radicals. It was highlighted that these reactions with $\text{OH}\bullet$ could create new radicals such as $(\text{Cl}\bullet^-, \text{Cl}_2\bullet^-$ and $\text{SO}_4\bullet^-)$, which are of low oxidation potential [108].

Owing to the existence of $\text{Cl}\bullet^-$ ions in the reaction, the production of $\text{Cl}_2\bullet^-$ radical anions can be evaded by monitoring the pH throughout the reaction time at a value of 3, which indicates that the degradation process of organic substrate through the photo-Fenton method aid in the formation of the acid consequently reduces the pH. This happens due to pH reduction (smaller than 2.5) in the existence of $\text{Cl}\bullet^-$ ions, resulting in more severe production of ferric chloride compounds (FeCl_2^+ and FeCl_2^+). These components also endure photolysis, thus reducing the volume of Fe(OH)^{+2} , which is the major supply of $\text{OH}\bullet$ in the photo-Fenton method. Furthermore the creation of the $\text{Cl}_2\bullet^-$ radical anion can

react with Fe^{2+} (reacting to Fe^{3+} with no production of OH^\bullet) and the organic substrate. One of the major benefits of utilizing the photo-Fenton method in comparison to the other oxidation methods is the application of solar radiation in its reaction process [109]. The existence of oxalate ions $(\text{C}_2\text{O}_4)^{2-}$ and the Fe^{3+} present in the reaction medium can produce the $[\text{Fe}(\text{C}_2\text{O}_4)]^+$ compound (Equation (19)) with light absorption at wavelengths lower than 570 nm [110,111]. The solar photoreactor setup is shown in Figure 4 by Pourehie and Saïen [111].

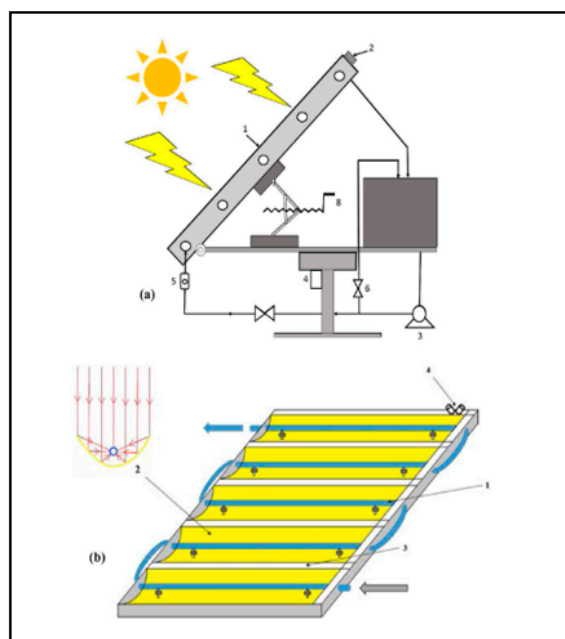
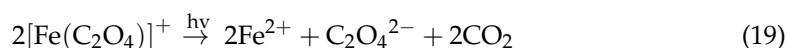


Figure 4. (a) The used solar photoreactor setup. (1) the surface of solar reactor, (2) light sensor, (3) recycle pump, (4) electromotor for rotation, (5) flow meter, (6) flow recycle valves, (7) storage tank, (8) and dynamic Jack. (b). The surface of the solar reactor; (1) quartz tubes, (2) parabolic polished aluminum reflector sheets, (3) Wind crossing paths, and (4) light sensor (Reprinted with permission from ref. [111]. Copyright 2020 Elsevier).



The ability to utilize solar radiation represents massive conservation from the energetic perspective, given that there is no necessity to utilize lamps. The progress of pilot plants for treating the wastewater that utilizes solar reactors used to the AOPs has been one of the major aims of study in the field [13,103].

The application of Fenton and photo-Fenton processes in the wastewater treatment process was studied by Giwa et al. [102] who use ferrioxalate as an iron source to analyze and reduce various wastewaters components, including BTEX and pentachlorophenol. Applying the photo-Fenton method has contrasted with additional photodegradation methods. Depending on the utilization of energy, the outcomes revealed that the method was above 30 times more effective than the $\text{UV}/\text{H}_2\text{O}_2$ and $\text{UV}/\text{Fe}^{2+}/\text{H}_2\text{O}_2$ methods. Therefore, the authors determined that the method utilizing ferrioxalate needs less electric energy compared to the $\text{UV}/\text{H}_2\text{O}_2$ process, leading to a decrease in the treatment costs.

The photo-Fenton method was applied to reduce the existing hydrocarbons in the produced wastewater synthesized using gasoline oil to mimic the water generated from oil industries in oil production fields that include dissolved salts with great amounts. It was noticed that the existence of NaCl (salts) prohibit the total degradation of contaminants. This was attained in the assessments without adding any amount of salt to the prepared wastewater [112]. The researchers assigned this impact to Fe^{3+} ions that reacted with Cl^- ions producing FeCl_2^+ composites, which, in the existence of UV radiation, can provide an

increase to $\text{Cl}_2^{\bullet -}$ radicals with smaller oxidation potential compared to HO^{\bullet} radicals, thus decreasing the productivity of the method. Fenton and photo-Fenton methods oxidation potential were examined to reduce different contaminants in the solution, including BTX and water polluted with gasoline including ethanol with a percentage of 25%. The degradation of prompt BTX composites was noted, along with the creation of transitional phenolic complexes in the primary reaction times that were degraded with 30 min of reaction. About 75% of total hydrocarbons introduced in gasoline-polluted water were removed and degraded, and the BTX composites were eliminated in the initial reaction mins (when phenolic composites were produced), after which they degraded sluggishly [113].

The impact of the photo-Fenton process in degrading phenol composites was examined by Brillas et al. 2020, which is an extremely popular contaminant in petrochemical production utilizing black light fluorescent lights as a source of UV-A emission. These are not expensive and are more efficient from the energy use perspective in comparison to medium-pressure mercury vapor lights. The effectiveness of black light fluorescent lamps was verified when compared to the degradation of phenol applying dark Fenton and photo-Fenton methods. The rate of degradation was twice as high when applying the photo-Fenton method. The degradation rate of phenol improved when the intensity of the radiation emission source increased, due to the increase in Fe^{2+} ion production (Equation (17)) [114]. Thus, this then increased the number of HO^{\bullet} radicals that were produced. These studies showed that the analysis of this factor is crucial as it facilitates the construction of the reactor that utilizes lamps as a source of UV radiation, which is essential for continuous wastewater treatment. This cannot be achieved by solar reactors only, which indicates that they are just used in cases of sunlight availability.

The application of the photo-Fenton method for the treatment of wastewater including diesel oil was examined by Hassan and his colleagues (2020). They performed research to enhance the number of chemicals utilized and to attain high degradation rates using a lesser number of chemicals. It was found that, by utilizing a small concentration of 0.1 mM Fe^{2+} ion, the rate of degradation of the organic composites existing in the wastewater was 90% classified as total organic carbon (TOC) [115]. The photo-Fenton method needs to be efficient from the commercial approach below these operating conditions, as it does not need successive treatment to eliminate the remaining iron. Besides, the degraded acid that exists in the wastewater from oil processing plants includes emulsified oil and a sequence of small-biodegradability micro contaminants, for example sulfides, phenols, ammonia, and mercaptans using different advanced oxidation process (AOP) methods.

Among the methods studied, Fenton and photo-Fenton achieved the most significant findings. The reaction of the dark Fenton process was applied until reaction stagnation with a separation efficiency for initial dissolved organic carbon (DOC) of only 27%, supported with the photo-Fenton method using UV radiation. The use of both methods together utilized constantly decreased the preliminary amount of DOC with a high percentage (more than 90%). The BTEX contaminants existing in the initial wastewater were eliminated to non-measurable amounts. The studies confirmed that the two methods were effective in eliminating contaminants from wastewater, but by utilizing high amounts of chemicals. They confirm that there is a need for optimizing the processes to decrease the number of chemicals used and thus increasing the efficiency of the process [116].

The productivity of Fenton chemicals in mineralizing organic complexes existing in water polluted with crude oil was also examined. The process was evaluated by observing the H_2O_2 using spectrophotometric analysis and via the CO_2 titrimetric quantification which allowed researchers to determine the level of mineralized TOC. These assessments indicated a 75% decrease in the initial TOC volume of water polluted by petroleum oil [117]. The efficiencies of the Fenton and photo-Fenton methods in the treatment of petroleum wastewater are summarized in Table 5.

Table 5. The efficiencies of Fenton and Photo-Fenton methods in the treatment of petroleum wastewater.

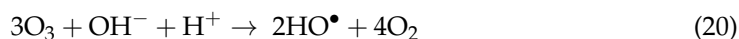
No	Method	Parameter	Removal Efficiency (%)	pH	H ₂ O ₂ (ppm)	Fe ²⁺ (ppm)	Ratio of H ₂ O ₂ :Fe ²⁺	Reaction Time (min)	Ref
1	H ₂ O ₂ /Fe ²⁺ /solar	TOC	84	2	1	0.08	12.5	300	Aljuboury et al. 2017
2	H ₂ O ₂ /Fe ²⁺	COD/ TOC	56 COD 54 TOC	4.3	9.7	1.1	8.8	120	Edison et al. 2019
3	H ₂ O ₂ /Fe ²⁺ /UV	COD	92	3	110	35	3.14	92	Majed et al. 2020
4	H ₂ O ₂ /Fe ²⁺ /solar	COD	84	3.2	200	1.5	133	180	Asaithambi et al. 2017
5	H ₂ O ₂ /Fe ³⁺	TOC	90	3	500	250	2	120	Deng et al. 2017
6	H ₂ O ₂ /Fe ²⁺ /UV	COD	72	5.6	17.86	1.76	10.14	70	Shokri et al. 2019
7	H ₂ O ₂ /Fe ³⁺ /TiO ₂	COD	69.6	3	1600	30	53	60	Hassan 2018
8	H ₂ O ₂ /Fe ²⁺ /UV	COD	76.8	3	250	40	6.25	30	Tufaner 2020

Table 5 shows the efficiencies of the Fenton and photo-Fenton process in the treatment of petroleum refinery wastewater and the separation of different contaminants including TOC and COD compounds. The studies showed that, at different conditions including the pH, reaction time, and H₂O₂ concentration, Fe⁺² concentrations under both processes were effective in eliminating contaminants from the wastewater but utilizing high amounts of reagents.

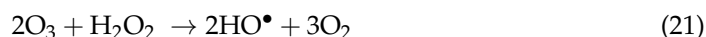
5.4. Ozone Based AOPs and Their Application in Petroleum Wastewater Treatment

The use of ozone (O₃) has been extensively applied to eliminate undesirable organic pollutants in both purification of drinking water and wastewater treatment [118]. Ozone (O₃) is a selective oxidant that reacts with electron-rich organic compounds at a high reaction rate constant (kO₃, pH7 > 105 M⁻¹ s⁻¹) [119,120]. Hydroxyl radical (OH•) produced from the consumption of O₃ is a nonselective oxidant, which quickly reacts with several organic composites at closely controlled rates of diffusion [121].

The creation of radicals, for example the superoxide radical (O²⁻•) and hydroxyl radical (HO•), happens when ozone is combined with water, via a complicated series of reactions. The ozone degeneration rate in water is improved at a greater pH [122]. The reaction integration with HO• and molecular ozone produces the oxidation of the organic compound.



The ozone decomposition cycle can be improved by adding hydrogen peroxide (H₂O₂) to cause the creation of HO• [123]. H₂O₂, to some extent, separates in water to generate the hydroperoxide ion. These HO²⁻ ions react with the O₃, promptly generating the HO•. In brief, the reactions integration results in the next equation:



Different parameters determine the performance of the process including the contact time, water alkalinity, and ozone concentrations. By presenting hydrogen peroxide (H₂O₂) when the highly reactive materials oxidized with O₃, it improves the degradation rate. The ozonation method using H₂O₂/O₃ systems was found to be the greatest proper AOP method in the purification of water in comparison to other technologies of AOPs. Therefore, there is a discipline-verified operation history in the application of H₂O₂/O₃ techniques. Another approach to accelerate ozonation is by utilizing catalysts (homogenous or heterogeneous). Research has been conducted with several metal ions, and metal oxides have demonstrated considerable variations in decomposition, but just in certain situations. The amount of ozone in the inlet gas and the ozone process time have been deemed as major parameters to develop the degradation rate. The research of oxidation has been conducted on numerous organic pollutants utilizing TiO₂, Fe₂O₃, Mn²⁺, MnO₂, Fe²⁺, and Fe³⁺. The system of ozone/catalyst was found to be more efficient for the COD and TOC reduction, and ozone oxidation only at a high pH [124]. In the tests conducted with [125,126], it was noticed that the chemical decomposition is successfully enhanced with the synergistic impact of photocatalytic reactions happening on TiO₂-photocatalyst. The experimental setup of the photoreactor is shown in Figure 5 [126].

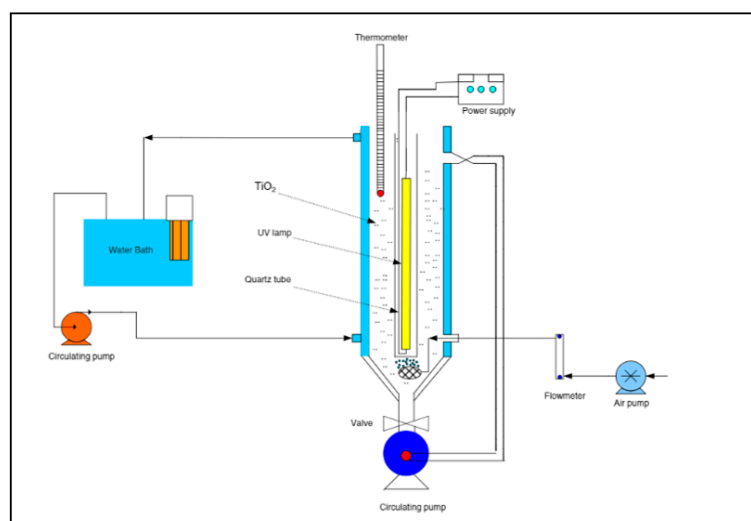
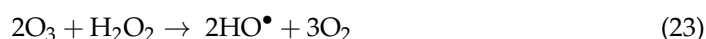
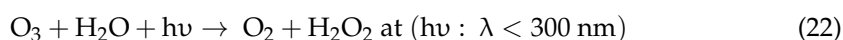


Figure 5. The experimental set up of photoreactor (Reprinted with permission from ref. [126]. Copyright 2012 Elsevier).

The ozone process can also be improved through the decomposition of ozone with direct absorption via UV radiation ($\lambda = 254 \text{ nm}$) which produces H_2O_2 as a transitional and then decomposes to $\text{HO}\bullet$ radicals. The reactions shown in Equations (22) and (23) demonstrate the photolysis of ozone to produce H_2O_2 , which leads to the creation of a very reactive $\text{HO}\bullet$ radical [127]. Though numerous oxidation methods can damage and eliminate organic pollutants, $\text{HO}\bullet$ is the main separation method.



This technique includes all the mechanisms of organic degradation via O_3/UV , $\text{H}_2\text{O}_2/\text{O}_3$ and $\text{H}_2\text{O}_2/\text{UV}$, O_3/Fenton , catalytic ozonation ($\text{O}_3/\text{Metal oxide}$), and photocatalytic ozonation, etc. [125]. The detailed steps of the mechanism of organic degradation of wastewater contaminants through O_3/UV are described in Figure 6. It is more efficient to separate organic materials than applying UV or O_3 separately, and it is an additional effective means of producing $\text{HO}\bullet$ in comparison to the $\text{UV}/\text{H}_2\text{O}_2$ technique at the same oxidant dosages [128,129]. However, this procedure is costly and energy-intensive [130]. Moreover, ozone techniques have the possibility to create bromate [131,132]. Inhibiting composites can influence the absorption of UV radiation, therefore decreasing the effectiveness of the method. Nevertheless, this difficulty will be lessened by increasing the ratio between the H_2O_2 and O_3 , decreasing the pH, or increasing the hydroxyl radical amount [133,134]. In addition, it is found that the absorption of UV radiation by O_3 was affected by the turbidity. The ozone production energy costs are the major operating expenses for this procedure. The existence of carbonate or bicarbonate may behave as hunters of the hydroxyl radical in normal structures which is an additional problem of these methods [135]. The usage of AOPs with ozone has more industrial uses in water treatment in comparison to all other AOPs. The method of ozone has been utilized to handle the petroleum refineries wastewater pollution.

A study conducted by [136] on the biochemical wastewater sewage treatment plant uses a combination of O_3/UV advanced oxidation process. The authors showed that the major parameters affecting the efficiency of the UV/O_3 process were the efficient O_3 distribution means, monitoring of the comparative pressure in the reactor, the ratio of ozone added, and UV radiation intensity. Cruz and his workers named the ozone concentration that consumes reactions with the initiator's composites as the ozone dose line, beyond which the addition of H_2O_2 rises the rate of $\text{HO}\bullet$ production. The authors

noticed that, in the case of H_2O_2 addition, ozonation does not have much effect on the exposure of $\text{HO}\bullet$ until the concentrations become more than the threshold ozone dosages used [137,138]. It was noted that the usage of a homogeneous reactor allows the quick and high formation of the radical species directly in the bulk liquid without any movement and mass transfer limits in comparison to a classical gas–liquid reactor, whereby the oxidation of the contaminants through hydroxyl radicals could be restricted via their transportation from the liquid phase to the interface as these radicals are extensively reactive and do not disperse in the solution [139]. The study carried out by Meshref et al. [140] on the identification and monitoring of the volatile organic compounds (VOCs) presents that the effluent’s wastewater showed 43% and 34% of COD and BOD reduction, respectively, resulting in an effective AOP process for VOCs degradation [141]. It also showed that, at certain ozone concentrations between 0.5–1.5 mg O_3 /mg dissolved organic carbon, the efficiencies of removing the contaminants increased up to ~14–18% in groundwater, above 6–10% in surface water, and insignificantly in secondary effluent through the $\text{O}_3/\text{H}_2\text{O}_2$ treatment in comparison to the conventional ozonation method.

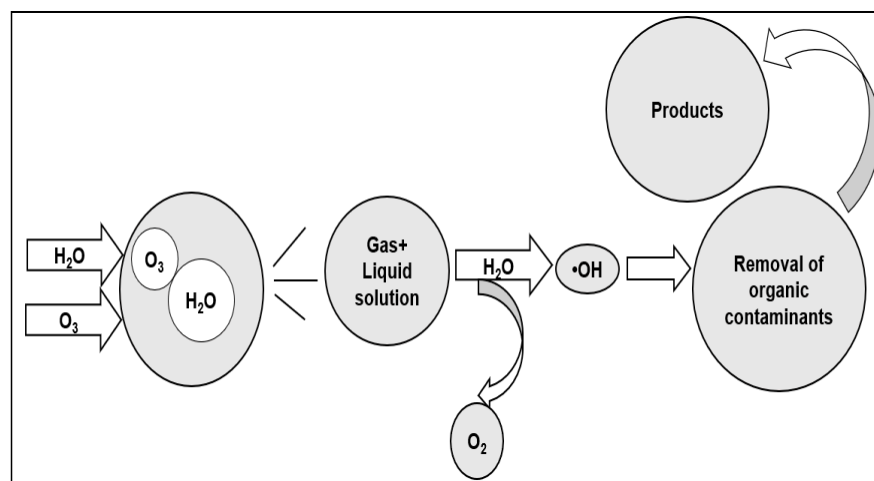


Figure 6. The detailed steps of the mechanism of organic degradation through the O_3 /UV method.

6. Integrated AOP Processes

In recent years, AOP methods have been extensively used for the treatment of petroleum wastewater applying different types of AOP methods. A combination of AOPs methods and additional conventional methods for wastewater treatment have been confirmed to be more efficient for the treatment of contaminated industrial wastewater sources efficiently. The selection means of these methods rely on the waste stream properties, environmental policies, and the cost.

AOPs were integrated with biological methods and other different processes to increase the efficiency of the AOP process in the degradation and separation of contaminants. Biological treatment methods include the decomposition of every residual oil, the degradation of organic composites, the breaking of organic contaminants, the removal of trace metals and nutrients, etc. [4,142]. Various methods of biological treatment have been effective in treating petroleum refinery wastewater. Between them, the activated sludge process (ASP) is the most biological of treatment methods used [143]. Ebrahimi et al. and Jain et al. [31,144] noticed a COD separation effectiveness ranging from 70–80% through the treatment of petroleum refinery wastewater utilizing ASP integrated with the AOP methods. A comparatively greater COD separation efficiency of 96% was achieved by Mirbagheri et al. [145], which was reported for the great synthetic aeration generated in the method. The integration of binary or extra-biological methods or the combination of the biological method with membrane-based AOP techniques were also found to be an efficient method for wastewater treatment. The study carried out by [146] combined an expanded granular sludge bed bioreactor (EGSB-BR) with an ASP-based AOP for petroleum

wastewater treatment, with a COD of 4600–5300 mg/L, which achieved a decrease in COD of 85% after a 62.8 h retention time. An application of biological-AOP was carried out by Razavi et al. [147], utilizing hollow fiber membrane bioreactors (HF-MBR) for the treatment of refinery effluent and attaining a COD separation of 82% after a 36 h retention time. The research reported by El-Naas et al. [148] utilized the anaerobic baffled reactor (ABR) for the treatment of produced water including heavy oil by applying a biological-based advanced oxidation process. The removal efficiency of 65% and 88% were then attained for heavy oil and oil and grease, respectively. The study reported by [149] examined the feasibility of combined electrocoagulation and membrane process. The combination of electrocoagulation and membrane filtration methods was used with integrated intelligent automation and process optimization in the structure of a mobile treatment platform (Figure 7). Up to 95% of oil, TOC, COD, TSS, and turbidity in produced water could be removed. The system could also effectively remove about 90% of ions, including chloride, sulfate, sodium, calcium, manganese, etc.

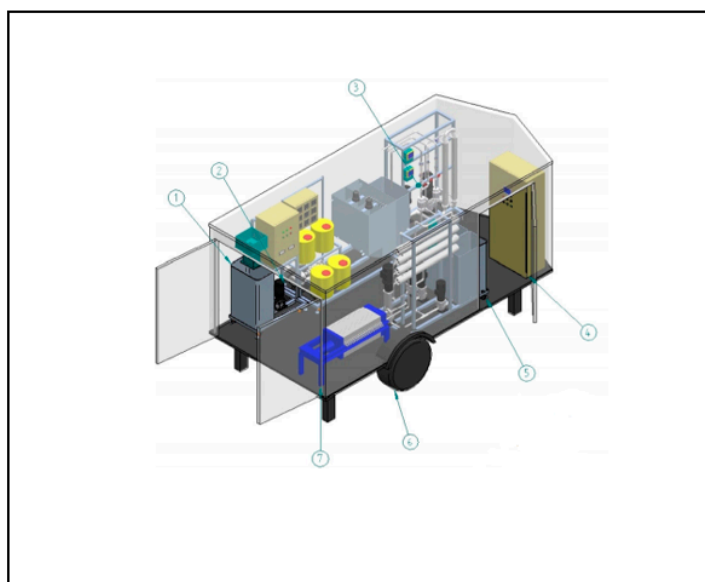


Figure 7. The integrated mobile system for oilfield produced water treatment. (1) EC equipment; (2) reaction/sedimentation equipment; (3) UF membrane unit; (4) controller; (5) RO unit; (6) trailer platform; (7) pressure filter (Reprinted with permission from ref. [147]. Copyright 2017 Elsevier).

However, most of these procedures generate a huge quantity of sludge, trained staff, and frequent maintenance. Certain methods also require extremely high hydraulic retention time (HRT), which is an essential drawback to the Bio-AOP techniques.

Another integrated advanced oxidation process was reported by [150]. They joined photo-bioreactors (PBR) with conventional oxic/anoxic methods for the treatment of petrochemical wastewater with an insignificant COD concentration of 312.8 mg/L and attained a separation rate of 71% using an electrocoagulation-based AOP method. A high concentration of petrochemical water with COD values in the range 3600–5300 mg/L was reduced and a separation rate of 97% was achieved in a spouted bed bioreactor (SBBR) with a packed activated carbon treatment (PACT) and an electrocoagulation cell (EC). There are several studies in integrating other AOPs, for example integration of photocatalysis and ultrasound [151], ozonation and ultrasound integration [152], the integration of Fenton, and photo-Fenton processes with ultrasound [153]. Fernandes et al. [151] showed that the integration of photocatalytic and electrochemical processes achieved a high degradation rate compared to the photocatalytic AOP process alone. The major factor in the integration methods to assess the efficiency of the system is the synergetic impact. The synergetic impact is a factor that indicates the development in the degradation of organic compounds below the integrated process. In general, the studies found that the integration of ad-

vanced oxidation technology with other methods is simple, effective, and economic for the treatment of wastewater, except when employing biological methods. The biological process is uncommon due to the creation of hydroxyl radicals throughout the AOPs which can negatively affect the biomass. Furthermore, the existence of H_2O_2 is also toxic to microorganisms. Consequently, it is better to utilize the integrated procedure in the AOP part to improve biodegradability and oxidation in a short time.

AOP-bioremediation is a new type of integrated method applied for industrial wastewater to separate micropollutants. The AOP-bioremediation integrated process concentrates on the treatment of petroleum industrial wastewater including bio-resistant pollutants. The combined two-step process has been studied, partially owing to the capability of AOPs for the degradation of these toxic pollutants to biodegradable compounds [103]. However, the recent literature from the last two years has indicated that the feasible application of the integrated process might be difficult, owing to the variety of potential effluent properties, operating conditions, and design structures. Consequently, the integrated process of AOPs with bioremediation has been recently studied for wastewater treatment [154].

AOP methods were also combined with the membrane (membrane-based AOP) as an innovative technique for the degradation of pollutants in the wastewater. This method is strongly related to the photocatalytic process which utilizes semiconductor catalysts [155,156]. A membrane gives an extra advantage of filtration. [157] This explains the mechanism of the process where the nanoparticle catalysts are inserted and dispersed in the reactor suspension in the form of soluble particles, and where the membrane performs as a barrier for the photocatalyst particles, i.e., the particles are settled inside or on membrane surface. The combination of photocatalyst nanoparticles is predictable to develop the quality of the permeate via the reduction oxidation process which generated active radicals to degrade the pollutants.

7. Knowledge Gaps and Future Perspectives

Hydrocarbon existing in petroleum are categorized as important contaminants. The contaminants which are introduced in wastewater of the petroleum industry can be efficiently removed by utilizing various techniques. Studies on the review of new techniques with the least environmental and economic impact showed that it is a research focus field. This review concentrates on reviewing AOP methods applied for petroleum wastewater treatment produced with the activities of the petroleum industry. The wastewater produced by the petroleum industry includes several harmful materials, for example xylene, toluene, ethylbenzene, benzene, phenols, polycyclic aromatic hydrocarbons (PAHs), etc. It is extremely challenging to immediately separate the contaminants through applying a single technique, especially biological treatment, which is deemed as a green method. Therefore, advanced oxidation process treatments in integration with other treatment techniques are needed. It has been described that the combination of different methods could provide greater outcomes more than a single process utilized for oily wastewater treatment. However, understanding the concept of technology combination is still at its initial stages which require to be investigated by scientists. The latest improvements in the process are mainly more costly, and need maintenance and long time. Future techniques require simple operation techniques which are appropriate for the petroleum industry and other different industries. A massive volume of solid and liquid waste is generated, owing to the activities of the petroleum industry. Controlling and treating the produced waste is currently a significant problem in any country. Owing to the increase in wastewater production, appropriate removal, treatment, and recycling are presenting extra problems, as treatment and disposal are extremely expensive. In addition, waste recovery is developing as a focus research field since it presents sustainability of the environment and social possibilities. Several studies are concentrating on the recovery of several supplies produced by human activities, for example energy, bio-products, metals, and oil from wastewater. Significant pollutants are introduced in petroleum industrial wastewaters, which can be considered as resources after recovery. Effective resource recovery and recycle can generate green life, while, on the other hand, it can help in developing economy through decreasing waste and enhance

environmental health and recovery cost. Consequently, there is a necessity to recover and recycle the produced waste from petroleum industry activities in an effective way. Viable treatment methods to generate less contaminated products generate a new approach for the sustainability of the environment and economy. To enhance the development of the petroleum wastewater industry, there is a necessity to create sustainable techniques such as AOP processes. The development of AOP methods can open up new chances for the market and attain effective application of resources.

Moreover, these technologies are favorable and valuable methods where toxic compounds are efficiently degraded and separated from the wastewater. Therefore, these methods were assessed at a pilot scale. Different combination with other types of studied AOP were studied at highly recyclable catalysts to evaluate the treatment efficiency of the wastewater before discharged with no hazardous impacts to the environment. In addition to the future studies on this method at pilot scale treatment unit, the studies must also contain the application of accessible AOPs based on modified catalysts, for example titanium dioxide, with an improvement in the process efficiency in visible light, following that a remarkable decrease in the process costs related to the energy consumption. However, the difference between the catalyst costs (modified catalysts) and energy costs for other types of AOP methods stays a wide-open case. AOP methods give a green, economic, and efficient methodology for the deep degradation of contaminants from petroleum refinery wastewater. Other future research viewpoints would be targeted near the use of some combined methods with AOP methods, such as fluidized-bed Fenton, fixed bed Fenton processes, etc., which study the competence of electro-Fenton processes for treating petroleum wastewater and identify the reaction kinetics and mechanisms that utilize newly developed catalysts as an effort to enhance the total oxidation productivity.

8. Conclusions

Petroleum wastewaters can be handled via the oxidation of chemical and biological treatment methods. There are various harmful components introduced in wastewater produced through the petroleum industry activities. An effective treatment approach should be designed to consider the environment and human health. AOP is one of the techniques which is getting a lot of worldwide concern for cleaning petroleum contaminants such as hydrocarbons. Nevertheless, the combination of different methods provide greater findings more than a single procedure utilized for the treatment of wastewater. The present wastewater treatment methods were concentrated to eliminate the contaminants from wastewaters to match the wastewater release criteria. To settle the risks related to the components of petroleum, an appropriate technique that handles the waste and removes those contaminants from wastewater would be a favorable choice. Recent studies related to the application of advanced oxidation processes (AOPs) in petroleum wastewater treatment that use the latest advanced oxidation types either separately or by combination with reverse to wastewater characteristics for generating non-toxic by-products were presented. A highly comprehensive literature review was presented for comparing the different AOP technologies described in this review, and a discussion about their advantages and disadvantages was also highlighted.

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Abbreviation

AOP	Advanced oxidation process
BTEX	Benzene, Toluene, Ethylbenzene Xylene
RFCC	Residual Fluid Catalytic Cracking
UV	Ultraviolet
BOD	Biological oxygen demand
COD	Chemical oxygen demand
PAHs	Polycyclic aromatic hydrocarbons
SBBR	Spouted bed bioreactor
PACT	Packed activated carbon
EC	Electrocoagulation cell
ABR	Anaerobic baffled reactor
ASP	Activated Sludge Process
HF-MBR	Hollow fiber membrane bioreactors
PBR	Photo-bioreactors
EGSB-BR	Expanded granular sludge bed bioreactor
VOCs	Volatile Organic Compounds
TOC	Total organic carbon
O ₃	Ozone
OH•	Hydroxyl radical
O ²⁻ •	Superoxide radical
H ₂ O ₂	Hydrogen peroxide
MnO ₂	Manganese dioxide
Fe ₂ O ₃	Iron III oxide
TiO ₂	Titanium dioxide
Fe ²⁺	Ferrous iron
Fe ³⁺	Ferric iron
Mn ²⁺	Manganese
Min	Minute
ppm	Part per million
Ref	Reference
DOC	Dissolved Organic Compounds
C ₂ O ₄ ²⁻	Oxalate ions
MTBE	Methyl tert-butyl ether
mg/L	milligram/litter
g/l	gram/litter
TPH	Total petroleum hydrocarbon
Ppb	Parts per billion
E ₀	Oxidation potential
CHR	Chrysene
BbF	Benzo[b]fluoranthene
USEPA	United States Environmental Protection Agency
VFA	Volatile fatty acids
TDS	Total dissolved solids
TSS	Total suspended solids
DO	Dissolved oxygen
HEM	Hexane Extractable Material
MBAS	Methylene Blue Active Substance
API	American Petroleum Institute
O & G	oil and grease
hr	hour

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