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Waste Water Treatment from Petrochemical Industries: The Concept and Current Technologies-A review

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معالجة ميــاه الصرف الناتجة من الصناعات البتروكيماوية: المفهوم والتقنيات الحالية – مراجعــة

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Abstract

Increasing consumption of oil in modern society has led to more oil refinery waste generation. The wastewater from these industries mainly contains oil, organic matter, and other compounds that need to be well managed before they can be discharged to any receiving waters. The treatment of this wastewater can be carried out by physical, biological, and chemical treatment processes. Treatment of petroleum wastewater has two stages, firstly, the pre-treatment stage reduces grease, oil, and suspended materials. Secondly, an advanced treatment stage is to degrade and decrease the pollutants to acceptable discharge values. The current review summarizes studies and investigations carried out for the treatment of petroleum industries and refineries

Keywords: *Effluent, Chemical Treatment, Oxygen demand, Physical Treatment.*

الملخص

يتطلب التزايد المستمر على الطاقة مزيدا من الاستكشاف واستغلال للموارد الطبيعية والتي تشمل النفط الخام المسئول الأول عن التلوث حيث أدى زيادة استهلاك النفط والصناعات البتروكيميائية (PRE) الناتجة عنه في المجتمع الحديث إلى توليد المزيد من نفايات مصافي النفط. تحتوي المياه العادمة الناتجة عن هذه الصناعات بشكل أساسي على المواد العضوية والمركبات الخطيرة الأخرى كالنفايات والتي تحتاج إلى إدارة جيدة قبل أن يتم تصريفها إلى أي مياه غير ضارة بالوسط البيئي. يمكن إجراء المعالجة لهذه المياه العادمة من خلال عمليات المعالجة الفيزيائية والبيولوجية والكيميائية. تنقسم معالجة المياه العادمة البترولية أو الناتجة عن هذه الصناعات إلى مرحلتين: أولاً: مرحلة ما قبل المعالجة لتقليل الشحوم والزيوت والمواد العالقة، وثانياً: مرحلة المعالجة المتقدمة لتخفيض الملوثات وتقليلها إلى قيم تصريف مقبولة. تلخص هذه الورقة بشكل علمي مختصر الدراسات والبحوث التي أجريت في هذا الصدد وكذلك النتائج التي تم التوصل إليها.

الكلمات الدالة: المعالجة الفيزيائية، الأكسجين اللازم لعمليات المعالجة، المعالجة الكيميائية.

1. Introduction

The treatment of wastewater generated by industrial activity is a major concern for plant operators and in particular those of refineries and petrochemical units. Every year a huge amount of wastewater is discharged directly into watercourse, causes the large environmental

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problems. Treating wastewater physically, biologically and chemically is vital for the nutrient cycling and maintaining the ecosystem stability

Petrochemical wastewater is a general term of wastewater associated with oil-related industries. The sources of petrochemical wastewater are diverse and can originate from oilfield production, crude oil refinery plants, the olefin process plants, refrigeration, energy unities, and other sporadic wastewaters. The compositions of wastewater from different sources consist of varying chemicals and show different toxicity and degradability in terms of biological treatment.

Oilfield-produced wastewater is generated in crude oil extraction from oil wells that contain high concentrations of artificial surfactants and emulsified crude oil characterized of high COD and low biodegradability. It is produced during oil extraction in oil fields and contains complex recalcitrant organic pollutants such as polymer, surfactants, radioactive substances, benzenes, phenols, humus, polycyclic aromatic hydrocarbons (PAHs), and different kinds of heavy mineral oil. Table (1) presents the commonly found compositions of wastewater obtained from oilfield production.

Table 1. Wastewater parameter form oilfield production (Benyahia *et al*., 2006).

Parameter	Values	Heavy metal	Values (mg/L)
Density (kg/m^3)	1014–1140	Calcium	13–25,800
Surface Tension (dynes/cm)	43–78	Sodium	132–97,000
TOC (mg/L)	0–1500	Potassium	24–4300
COD (mg/L)	1220	Magnesium	8–6000
TSS (mg/L)	1.2–1000	Iron	< 0.1–100
pH	4.3–10	Aluminum	310–410
Total oil (IR; mg/L)	2–565	Boron	5–95
Volatile (BTX; mg/L)	0.39–35	Barium	1.3–650
Base/neutrals (mg/L)	< 140	Cadmium	< 0.005–0.2
(Total non-volatile oil and grease by GLC/MS) base (g/L)	275	Chromium	0.02–1.1
Chloride (mg/L)	80–200,000	Copper	< 0.002–1.5
Bicarbonate (mg/L)	77–3990	Lithium	3–50
Sulfate (mg/L)	< 2–1650	Manganese	< 0.004–175
Ammoniacal nitrogen (mg/L)	10–300	Lead	0.002–8.8
Sulfite (mg/L)	10	Strontium	0.02–1000
Total polar (mg/L)	9.7–600	Titanium	< 0.01–0.7
Higher acids (mg/L)	< 1–63	Zinc	0.01–35
Phenols (mg/L)	0.009–23	Arsenic	< 0.005–0.3
VFA's (volatile fatty acids) (mg/L)	2–4900	Mercury	< 0.001–0.002
		Silver	< 0.001–0.15
		Beryllium	< 0.001–0.004

The composition of effluent in refinery wastewater depends on the crude quality. It varies with the operating conditions (Benyahia *et al*., 2006). In the refinery, non-hydrocarbon substances are removed and the oil is broken down into its various components and blended into useful products. So, petroleum refineries produce large volumes of wastewater including

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oil well produced water brought to the surface during oil drilling, which often contain a recalcitrant compounds and rich in organic pollutants therefore cannot be treated easily and difficult to be treated biologically (Vendramel *et al*., 2015; and Asatekin *et al*., 2009).

Removal of pollutants produced by industrial plants is requirement for reuse of water and obtains to environmental standards (Al-Meshragi *et al.*, 2009; Farajnezhad and Gharbani, 2012; and Aboabboud *et al.*, 2013). Petroleum wastewater are a major source of aquatic environmental pollution and are wastewater originating from industries primarily engaged in refining crude oil, manufacturing fuels and lubricants (Wake *et al*., 2005) and petrochemical intermediates (Harry *et al*., 1995). Coelho *et al*. (2006) reported that the volume of petroleum wastewater generated during processing is 0.4–1.6 times the amount of the crude oil processed. If the petroleum wastewater, which contained high organic matter, discharged into the aquatic environment, which required 2 mg/L from dissolved oxygen for normal life, results in decreased dissolved oxygen by the bacteria (Attiogbe *et al*., 2007). In anaerobic systems, the products of chemical and biochemical reactions produce displeasing colors and odors in water. So, the oxygen availability is important in water to reduce that (Attiogbe *et al*., 2007).

Various environmental protection agencies set maximum limits of discharge for each component of the waste as shown in Table (2) to protect environment from the hazardous composition in petroleum wastewater. The fuel additives, which were carcinogenic such as dichloroethane (DCE), Dichloromethane (DCM) and t-butyl methyl ether (tBME), were considered the most of un-degraded total petroleum hydrocarbon (Diya'uddeen *et al.,* 2011; and Squillance *et al*., 1996).

Table 2. Minimum standard discharge limits for refinery effluents

Composition (mg/L)	References		
	Ma *et al.* (2009)	**Environmental Health Safety Guidelines (2009)**	**Aljuboury *et al.* (2015a)**
COD	100	150	150-200
BOD	10-15	30	
TDS	-	-	1500-2000
SS	70	30	
TOC	-	-	50-75
Ammonia	15	-	-
Phenols	-	-	-
Sulphides	-	1.0	-
pH parameter	6-9	6-9	6-9

2. Current Petroleum Wastewater Treatment Techniques

The petroleum wastewater treatments are classified into three types; physical, chemical and biological. However, the treatment required a typical application of the integrated system due to the complexity of characteristics of petroleum wastewater. Thus, the conventional treatment methods need multistage process treatment. The first stage consisted of pre-treatment, which includes mechanical and physicochemical treatments followed by the second stage which is the

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advanced treatment of the pretreated wastewater. Based on the literature review conducted, the techniques and methods for petroleum wastewater treatment included physical, chemical, biological treatment processing.

2.1. Physical Treatment

Physical treatment methods include processes where no gross chemical or biological changes are carried out and strictly physical phenomena are used to improve or treat the wastewater. Examples would be coarse screening to remove larger entrained objects and sedimentation. The presence of sulphide and salts could inhibit biological operation in excess of 20 mg/L (Altaş and Büyükgüngör, 2008). Thus, the physical treatment system is a primary treatment step, which is essential to remove or separate suspended solids (SS), immiscible liquids, solid particles, suspended substances (Sancey *et al*., 2009) from petroleum wastewater by using sedimentation, coagulation and flocculation and prolonged use of the secondary treatment unit. Most physical treatment techniques are considered as conventional methods .

Nowadays, physical technologies such as sedimentation are used prior to biological treatment in order to remove suspended solids. The sedimentation treatment, which is used to separate oil from water, is mechanically achieved by gravity in API separators or separation tanks. Coagulation process was used to remove turbidity and organic load abatement. However, physical processes were relatively ineffective for the treatment of petroleum wastewater because of its complexity and therefore, other processes might be used for pretreatment.

As shown in Table (3), Wang *et al.* (2015) reported that the maximum reductions for total naphthenic acids (NAs) and aromatic naphthenic acids by the physicochemical processes were 16% and 24%, respectively in a refinery wastewater while they were 65% and 86% respectively, by the biological processes.

Conventional approaches to treating oily wastewaters have included gravity separation and skimming, dissolved air flotation, de emulsification, coagulation and flocculation. Gravity separation followed by skimming is effective in removing free oil from wastewater. Oil–water separators such as the API separator have found widespread acceptance as an effective, low cost, primary treatment step.

The API oil–water separator is designed to separate the oil and suspended solids from their wastewater effluents. The name is derived from the fact that such separators are designed according to standards published by the American Petroleum Institute.

The API separator, however, is not effective in removing smaller oil droplets and emulsions. Oil that adheres to the surface of solid particles can be effectively removed by sedimentation in a primary clarifier. Dissolved air flotation (DAF) uses air to increase the buoyancy of smaller oil droplets and enhance separation. Emulsified oil in the DAF influent is removed by de-emulsification with chemicals, thermal energy or both. DAF units typically employ chemicals to promote coagulation and increase flock size to facilitate separation.

Table 3. Overview of work done in the area of physicochemical treatment applications to treat the petroleum wastewater reported by various researchers

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No	The method applied	The wastewater type	Removed pollutants	Max. Removal efficiency (%)
1.	The physicochemical processes.	A refinery wastewater	Total naphthenic acids (NAs)	16
			Total naphthenic acids (NAs)	24
2.	An immersed membrane process.	Petroleum refinery wastewater	wastewater oil content	69
3.	Membrane bioreactor (MBR).	Petrochemical wastewater	Heavy metals	70
			Iron	75
4.	Nanocomposite membrane with the multi-walled carbon nanotube (MWCNT) incorporated in Polyvinylidene fluoride (PVDF) matrix.	refinery wastewater	Oil	
5.	A cross-flow membrane bioreactor (CF-MBR)	Petroleum wastewater	COD	93
6.	The hollow-fiber membrane bioreactor (HF-MBR)	Real petroleum refinery wastewater	COD	82
			BOD_5	89
			TSS	98
			VSS	99
			Turbidity	98
7.	Membrane sequencing batch reactor.	A synthetic petroleum wastewater	Hydrocarbon pollutants	97
8.	Ultra-filtration (UF) membranes.	Refinery wastewater	COD	44
9.	Poly aluminum chloride and ferric chloride for coagulation treatment.	Petroleum wastewater	COD	58
10.	Poly-zinc silicate (PZSS) and anion polyacrylamide (A-PAM) for coagulation /flocculation treatment.	Heavy oil wastewater	Oil	99
11.	Subsequent coagulation/H_2O_2.	Petroleum refinery wastewater	COD	58
			BOD5	78
12.	Coagulation by alum Coagulation by ferric chloride ($FeCl_3$).	Petrochemical wastewater.	COD	52
13.	Electro-coagulation.	Petrochemical wastewater	Phenol	100
14.	A cell with horizontally oriented aluminum cathode and a horizontal aluminum screen anode at high current density	Oil refinery waste effluent	Phenol	97
15.	Adsorption by organoclay.	Petroleum wastewater	organic substances	62

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16.	An activated carbon adsorption.	Petroleum wastewater	COD	60
17.	A microwave-assisted catalytic wet air oxidation process.	Petroleum wastewater	COD	90
18.	The $O_3/UV/TiO_2$ process.	Petroleum wastewater	Phenol	99.9
			Sulfide	97.2
			COD	89.2
			Oil	98.2
19.	Partial precipitant [$FeCl_3{\cdot}6H_2O$, and $FeSO_4{\cdot}7H_2O$] and coagulant aids [$Ca(OH)_2$ and $CaCO_3$]	Petroleum refinery wastewater	COD	75
			Sulfide	99

Emulsified oil in wastewater is usually pre-treated chemically to destabilize the emulsion followed by gravity separation. The wastewater is heated to reduce viscosity, accentuate density differences and weaken the interfacial films stabilizing the oil phase. This is followed by acidification and addition of cationic polymer/alum to neutralize negative charge on oil droplets, followed by raising the pH to the alkaline region to induce flock formation of the inorganic salt. The resulting flock with the adsorbed oil is then separated, followed by sludge thickening and sludge dewatering.

2.2. Coagulation Flocculation

Most wastewater treatment plant includes sedimentation in their process. The sedimentation also called clarification is a treatment process in which the velocity of the water is lowered below the suspension velocity and the suspended particles settle out of the water due to gravity. Settled solids are removed as sludge, and floating solids are removed as scum. Wastewater leaves the sedimentation tank over an effluent weir to the next step of treatment .

The efficiency or performance of the process is controlled by: retention time, temperature, tank design, and condition of the equipment. However, without coagulation/flocculation, sedimentation can remove only coarse suspended matter which will settle rapidly out of the water without the addition of chemicals. This type of sedimentation typically takes place in a reservoir, sedimentation or clarification tank, at the beginning of the treatment process.

Coagulation-flocculation consists on the addition on the clarification tanks of chemical products that accelerate the sedimentation (coagulants). The coagulants are inorganic or organic compounds such as Aluminum sulphate, Aluminum Hydroxide chloride or high molecular weight cationic polymer. The purpose of the addition of coagulant is to remove almost 90% of the suspended solids from the wastewater at this stage in the treatment process.

2.3. Adsorption Techniques to Treat Wastewater

Adsorption is a natural process by which molecules of a dissolved compound collect on and adhere to the surface of an adsorbent solid. Adsorption occurs when the attractive forces at the carbon surface overcome the attractive forces of the liquid. Granular activated carbon is a

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particularly good adsorbent medium due to its high surface area to volume ratio. One gram of a typical commercial activated carbon will have a surface area equivalent to 1,000 m^2.

An activated carbon adsorption is effective in removing organic compounds residual after biological treatment. In addition, low molecular weight pollutants are specially adsorbed (Lorenc and Gryglewicz, 2007). This method is limited by high consumption of activated carbon or the requirement for frequent regeneration of columns (Renou *et al*., 2008). El-Naas *et al*. (2009) achieved 30% COD reduction at the ambient temperature, whereas at 60 °C and 53% COD reduction was reached.

2.4. Membrane Technology

Membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are increasingly being applied for treating oily wastewater. Of the three broad categories of oily wastes –free-floating oil, unstable oil/water emulsions, and highly stable oil/water emulsions– membranes are most useful with stable emulsions, particularly water soluble oily wastes. Free oil, on the other hand, can be readily removed by mechanical separation devices which use gravitational force as the driving force.
Unstable oil/water emulsions can be mechanically or chemically broken and then gravity separated. Pre-treatment to remove large particles and free oil is needed, especially if thin-channel membrane equipment is used.

Membranes have several advantages, among them: (1) The technology is more widely applicable across a wide range of industries; (2) The membrane is a positive barrier to rejected components. Thus, the quality of the treated water (the permeate) is more uniform regardless of influent variations. These variations may decrease flux, but generally does not affect quality of its output, (3) No extraneous chemicals are needed, making subsequent oil recovery easier, (4) Membranes can be used in-process to allow recycling of selected waste streams within a plant, (5) Energy costs are lower compared to thermal treatments, and (6) The plant can be highly automated and does not require highly skilled operators. The chemical nature of the membrane can have a major effect on the flux. For example, free oils can coat hydrophobic membranes resulting in poor flux (emulsified oil is usually not as much of a problem, unless it is concentrated to such a high level that the emulsion breaks, releasing free oils).
Hydrophilic membranes preferentially attract water rather than the oil, resulting in much higher flux. Hydrophobic membrane can be used, but usually in a tubular configuration that allows a high degree of turbulence (cross–flow velocity) to be maintained to minimize oil wetting of the membrane.

Membrane processes have some limitations: (i) Scale-up is almost linear above a certain size. Thus capital costs for very large effluent volumes can be high, and (ii) Polymeric membranes suffer from fouling and degradation during use. Thus they may have to be replaced frequently, which can increase operating costs significantly. In spite of the above disadvantages, membrane processing of oily wastewaters, sometimes in conjunction with other methods for treating the residuals, is a commercial success with more than 3,000 polymeric UF/MF installations and over 75 inorganic/ceramic units worldwide.

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2.5. Biological Treatment

Biological treatment incorporates actions of different microbes to eliminate organics and stabilize hazardous pollutants in petrochemical wastewater. Stringent environmental standards and recycling of water for reuse have shifted focus to biological treatments because of its cost and pollutant removal efficiency. As the nature of petrochemical wastewater is very complex, biological treatment to remove pollutants still has challenges despite immense potentials.
Biological treatment is the use of microbial metabolism, so that the water was dissolved, colloidal organic pollutants into harmless substances are stable (Kriipsalu *et al*., 2007; and Sirianuntapiboon and Ungkaprasatcha, 2007). Currently handles more mature technology and is used frequently in activated sludge and biological filter methods. Activated sludge in the aeration tanks uses the current state vector as purifying microorganisms, by adsorption, and concentrated on the surface of the activated sludge microorganisms to decompose organic matter. The biofilter biological filter method is inside, so that the micro-organisms are attached to the filter, waste water from the top go down through the filter surface during adsorption of organic pollutants and decomposition by microorganisms will be destroyed .

Generally, biological treatment methods can be divided into aerobic and anaerobic methods, based on availability of dissolved oxygen (Zhao *et al*., 2006). In anaerobic systems, the products of chemical and biochemical reactions produce displeasing colors and odors in water. Thus, the oxygen availability was important in water to reduce displeasing colors and odors (Attiogbe *et al*., 2007).

2.6. Anaerobic Biological Process

Anaerobic digestion is a biological process in which bacteria break down organic matter in the absence of oxygen. Anaerobic biological treatment has excellent organic removal efficiency and an economical cost. Organic matter is converted into CO_2 and CH_4, and sludge during anaerobic biological treatment.

Anaerobic digestion has the advantages of producing methane as a renewable energy, requiring less space and having lower sludge generation than aerobic process. A literature review of anaerobic digestion on the petrochemical wastewater is given in Table (2). Petrochemical wastewater treated in anaerobic baffled reactor (ABR), sequence batch, and up-flow sludge blanket reactor (UASB) was commonly applied. It shows that organics in the petrochemical wastewater could be partially anaerobic digested at a removal efficiency depending on the chemical constituents, reactor type, operational conditions (temperature, loading rate, etc.), and wastewater sources. COD removal efficiency is used here as a general parameter to assess the performance of different systems. Crude oil extraction of light, medium, and heavy petroleum wastewater treatment by different anaerobic digestion systems at mesophilic or thermophilic conditions showed that in batch test over 56–71% COD removal was achievable at thermophilic condition (Table 2), while UASB system can achieve over 93% COD removal at mesophilic conditions for wastewater from light petroleum extraction (Table 2). It seems light petroleum extraction wastewater was generally easily degradable (over 71–93% removal) compared to the medium and heavy oil extraction wastewater. The setup of plug

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flow pattern and granular sludge application in UASB might also enhance the interaction between wastewater and organisms, giving higher efficiency. The removal efficiency decreases as the loading rate increases, indicating the inhibition effects to the organisms. Medium- and heavy oil-produced wastewater treatment efficiency was relatively low. Batch system gives generally better treatment efficiency for these two wastewaters at about 50–60% removal (Table 2), while UASB shows low efficiency at around 20–30% removal efficiency. The effects of toxic chemicals in the wastewater and high content of large organic molecules can be the reason for low efficiency.

2.7. Aerobic Biological Processes

Aerobic process has been applied widely in petrochemical wastewater treatment attributed to its features of easy operation, less sensitiveness to toxic effects, higher organisms' growth rate, etc. than the anaerobic system. Different aerobic reactors such as traditional active sludge, contact stabilization active sludge, sequence batch reactor (SBR) that applies active sludge and biological aerated filter (BAF), membrane bioreactor (MB), moving bed biofilm reactor (MBBR), aerobic submerged fixed-bed reactor (ASFBR) that applies biofilm, etc. have been tested to treat petrochemical wastewater from varying sources and presented in Table (4). Generally higher COD and chemical removal efficiencies by aerobic process are achieved than the anaerobic processes (Tables 2 and 3).

Table 4. Overview of work done in the area of Biological treatment applications to treat the petroleum wastewater reported by various researchers

No	The method applied	The wastewater type	Removed pollutants	Max. Removal efficiency (%)	Reference
1.	The reactor immobilized with microorganisms.	Petroleum refinery wastewater	TOC	78	Zhao *et al.* (2006)
			Oil	94	
2.	The aerobic biological process.	Petroleum wastewater	COD	86	Satyawali and Balakrishnan (2008)
3.	The anaerobic treatment process (a UASB reactor).	Petroleum refinery wastewater	COD	82	Gasim *et al.* (2013)
4.	The up-flow anaerobic sludge bed (UASB) reactor.	Heavy oil refinery wastewater	COD	70	Wang *et al.* (2016)
			oil	72	
			NH_3-N	90.2	
			COD	90.8	
5.	The up-flow anaerobic sludge blanket (UASB) reactor and a two-stage biological aerated filter (BAF) system.	Heavy oil wastewater	oil	86.5	Zou (2015)

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6.	A combined UASB and anaerobic packed-bed biofilm reactor.	Petroleum wastewater	COD	81.07	Nasirpour *et al.* (2015)
7.	The activated sludge system.	Petroleum wastewater	Naphthenic Acids (NAs)	73	Wang *et al.* (2015)
8.	The sequencing batch reactor system.	Petroleum wastewater	Phenols	98	Al Hashemi *et al.* (2015)
9.	Anaerobic submerged fixed-bed reactor (ASFBR).	Petroleum wastewater	COD	91	Vendramel *et al.* (2015)
			TSS	92	

3. Conclusion

Petroleum refinery effluents (PRE) are hazardous compounds containing waste. The discharge of these waste waters into the environment adversely affects the ecosystem. An increasing global energy demand requires greater exploration and exploitation of the raw material, crude oil, which is responsible for these pollutants.

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