

Technology for Treating Oily Wastewater Derived from Various Industries: A Review Paper

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ABSTRACT

The increasing number of industrial sectors has resulted in more and more contaminated water and wastewater containing various harmful pollutants including oil compound. Less treated or untreated oily wastewater could give a serious threat to both human health and environmental sustainability. Therefore, the appropriate treatment methods such as membrane filtration, coagulation/flocculation, biological treatment, adsorption, and relevant integrated methods have to be implemented well to enhance the quality of those wastewaters before they are flowed into the final disposal place. This effort will endorse the success of waste management and circular economy system by recycling unwanted industrial by-product including wastewater, so it can be reused for further productive purposes.

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1. Introduction

Industrial development has contributed directly and indirectly to the increase in oily wastewater production. This unwanted by-product refers to oil and water mixture in any composition along with other harmful contaminants, so the water cannot be used any longer for productive process unless it is treated properly to reach certain standard of characteristics [1]. Generally, apart from other contaminants, wastewaters produced by oil manufacturer and other related industries contain some fats, waxes, or mineral oil solution endangering the environmental sustainability as well as human health to some extent [2, 3].

Theoretically, oil can be classified into some forms including petroleum oil, mineral oil, animal oil, and vegetable oil or plant-based oil [1]. There are various industries produce oily wastewaters in different quality levels. Petroleum refinery company, edible oil manufacturers, food and beverage, restaurant industry, and cooling and heating industries are several prominent industries that could generate a huge amount of oily wastewater resulted from some processes like production, refining, storage, and transportation [2, 3].

2. Oily wastewater and industry

Even though the industrial wastewater itself is a challenge for protecting environment, the growing industry is important for both economic growth and human life. Edible oil, for example, has been an important part of human life for being used in many purposes like cooking, cosmetic, and pharmacy products. Edible oil industries such as palm oil, canola oil, and olive oil result in oily wastewater containing harmful contaminants that need to be regulated in terms of its acceptable discharge standard before its final disposal to the final open place [3].

2.1. Palm oil industry

Palm oil producers are growing fast in several countries such as Indonesia, Malaysia and Nigeria resulting in challenging environmental issues to the countries due to high organic and non-organic contaminants in palm oil mill effluent (POME) [4]. POME as a by-product of palm oil manufacturing process is thick viscous liquid waste having foul odor with high concentration of COD, BOD and oil and grease [4, 5]. Therefore, governments or other related institutions have to standardize the POME discharge. Table 1 illustrates the example of allowable values of POME to be discharged to the open waters [4].

Table 1. POME characteristics and acceptable discharge standard

Parameters	Raw POME	POME discharge standard
Temperature (°C)	80-90	45
BOD (mg/L)	25,000	100
COD (mg/L)	50,000	-
Oil (mg/L)	4,000	50
Total Solids (mg/L)	40,500	-
pH	4.7	5 - 9
Total Kjehdal Nitrogen (mg/L)	750	200

2.2. Canola oil industry

Canola oil made from rapeseed plant is widely used for cooking oil and as an additive substance for candles, inks and medicinal products [5]. In its production process, the rapeseed plant is slightly heated, crushed and extracted using hexane, then refined using water precipitation and organic acid to remove gums, and then deodorized by steam distillation. Despite having lower values of COD, TOC, suspended solid and oil and grease, canola oil effluent still has to be purified before its final disposal. A study which investigated canola oil effluent treatment reported some characteristics of raw canola oil wastewater as stated in table 2 [6].

Table 2. Characteristics of raw canola oil wastewater

Parameter	Range
COD (mg/L)	330
TOC (mg/L)	90
Oil (mg/L)	250
pH	9

2.3. Olive oil

Olive oil is mostly produced by manufacturers located in the Mediterranean countries. This oil is known as the healthiest cooking oil as it offers some advantageous points in terms of health perspective. Other than that, olive oil is also useful for cosmetics production and other personal needs such as soap and shampoo as it is rich in antioxidant elements. However, its oily wastewater could contain high level of harmful pollutants, as illustrating in Table 3, yielding poor quality of the wastewaters [7, 8].

Table 3. Characteristics of raw olive oil mill wastewater

Parameter	Range
pH	4 - 5
TOC (mg/L)	1800 - 3200
COD (mg/L)	129000 - 183000
Conductivity (mS/cm)	16 - 32
Oil (mg/L)	5200 - 7800
Total phosphorus (mg/L)	350 - 830

2.4. Petroleum industry

To date, petroleum industry valorizing crude oil to become higher valuable fuel products for various applications through refining process has been the main source of industrial oily wastewater. This is because, apart from the effort of creating alternative green energy sources, crude oil and other fossil fuel still act as the main source of energy. This phenomena has resulted in a massive amount of petroleum refinery wastewater having detrimental influence on both environment and

living creatures due to its toxic contents like oil, hydrocarbons, sulfides, ammonia and large quantities of inorganic salts [9] as stated in Table 4 [10-12].

Table 4. Characteristics of raw petroleum refinery wastewater

Parameters	Average discharge values	Acceptable discharge standard
COD (mg/L)	3340	140
BOD (mg/L)	280	17
Oil (mg/L)	200	23
pH	10.6	6.2
Temperature (°C)	41	22
Sulfide (mg/L)	38	0
Phosphorus (mg/L)	97	0

2.5. Restaurant industry

Restaurant industry cannot be separated from urban development as it is main part of modern lifestyle for the community. This industry can have a significant amount of oily wastewater containing high oil and grease particles generated from its operational activities such as washing kitchen utensils, cleaning and cooking food materials [13]. In many developing countries, the oily wastewater generated by restaurant industry mostly flows into foul sewers. It would lead to public sewage treatment plants that can cause drain pipes blockage, bad odor, and harmful open waters if left less or untreated [14]. Table 5 illustrates the common characteristics detected in some restaurant wastewaters [13].

Table 5. Characteristics of raw restaurant oily wastewater

Parameters	Range
COD (mg/L)	750 - 6800
BOD (mg/L)	600 - 2500
Oil (mg/L)	500 - 4700
pH	6.1 - 8
Suspended solid (mg/L)	250 - 650
NH ₂ -N (mg/L)	4.8 - 10.4

3. Existing Technology for Oily Wastewater Treatment

The untreated or less treated oily wastewater can endanger public health and ecological systems due to its hazardous components. Other than oil and grease, oily wastewaters may also contain suspended and dissolved solids, phenol, heavy metals, dyes, and other harmful particles [15]. Therefore, proper treatment processes for oily wastewater are needed to produce wastewater that meets the regulatory standards. Several existing technologies can be implemented to increase the quality of those oily wastewaters such as air flotation, membrane filtration, advanced oxidation, and adsorption.

3.1. Air flotation

For oily wastewater treatment, this method can be applied for reducing the amounts of fat, oil and grease by bubbling air through wastewater tank or pond in order to increase the specific gravity difference between pollutant particles and water [16]. In this process, suspended particles and oil globules can be floated to the wastewater surface to be removed by further skimming. There are some techniques of air flotation like dissolved air flotation (DAF), dispersed air flotation (DispAF) and electro-flotation. For DAF, wastewater is saturated under pressure with air generated by granular media filtration. Then, the pressure is released through needle valves into flotation cells resulting in air bubbles having about 30 - 120 µm in diameter [17]. For DispAF technique, air bubbles are produced through the pores of sintered glass disks with diameter ranging from 75 to 655 µm [18], while electro-flotation process involving electrolysis of water used with bubbles that is produced at the electrodes having diameters ranging from 22 to 50 µm [19].

3.2. Membrane separation process

Membrane separation is an advanced filtration process mainly driven by the pressure difference as an average value of inlet and outlet pressure, named trans membrane pressure. Other parameters

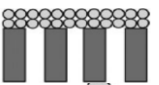


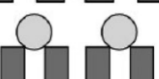
that affect the effectiveness of membrane filtration processes are cross flow velocity, temperature, pH, oil, and salt concentration [20, 21]. There are several types of membrane that commonly applied in wastewater treatment plants as described in Table 6 [22].

Table 6. Membrane characteristics

Membrane type	Structure	Driving force	Mechanism	Material	Polar Tendency
Electrodialysis	<i>Electrostatically charged membrane</i>	Electrical potential	Electrostatic diffusion	Styrene/ Vinylpyridene, Divinyl benzene-	-
Microfiltration	Symmetric microporous (0.02 - 10 μ)	Pressure, 1 - 5 atm	Sieving	Polypropylene, Polyethylene, Polycarbonate, Ceramic	Non polar
Ultrafiltration	Asymmetric microporous (1 - 20 nm)	Pressure 2 - 10 atm	Sieving	Polysulfone, Dynel, Cellulose acetate, Polyvinylidene fluoride	Non polar
Nanofiltration	Asymmetric microporous (0.01 - 5 nm)	Pressure 5 - 50 atm	Sieving	Polyvinylidene fluoride	Polar
Reverse osmosis	Asymmetric homogeneous skin, microporous support	Pressure 10 - 100 atm	Solution diffusion	Cellulose acetate, Polyamide, Nylon	Polar

Membrane filtration has some excellent points such as high contaminant removal percentage, automatic operation system, molecule separation based on manageable size dismissal, no specific chemical needed, and moderate skilled operators [23-26]. However, high loading oily wastewaters can lead quickly to membrane fouling caused by oil and other pollutant particles depositing onto membrane surface or membrane pores. When this happens, the performance of the membrane will decrease leading to less and less volume of permeate. Some models can be used to describe the fouling mechanism occurring on membrane pores during filtration. Hermia's models consisting of cake formation, intermediate pore blocking, standard pore blocking and complete pore blocking model are well acceptable models used for fouling mechanism analysis as described in Table 7 [27-30].

Table 7. Hermia's models and blocking mechanism

Blocking mechanism	n -value	Fouling mechanism illustration	Hermia's models
Cake layer formation	0		$\frac{1}{J^2} = \frac{1}{J_0^2} + K t$
Intermediate pore blocking	1		$\frac{1}{J} = \frac{1}{J_0} + K A t$
Standard pore blocking	1.5		$\frac{1}{J^{0.5}} = \frac{1}{J_0^{0.5}} + K t$
Complete pore blocking	2		$\ln(J) = \ln(J_0) - K t$

3.3. Adsorption process

Adsorption is the adhesion process of atoms, ions or molecules from a gas, liquid, or dissolved solid to adsorbent surface when they are in contact for specific time [31]. This process could take place through physical and chemical ways. While physical adsorption is the existence of inter-particle bonds between adsorbate and adsorbent reversibly, chemical adsorption experiences stronger ion or molecule aggregation by electron exchange irreversibly.

Activated carbon (AC) is one of the most prominent types of practical adsorbent used in adsorption process. However, the regeneration cost of the saturated AC is relatively expensive. This condition has opened the curiosity to explore a variety of low cost adsorbents derived from some agricultural and other organic wastes as displayed in Table 8. Those natural organic materials have several benefits including better economic value, simple usage, reliable availability, and high biodegradation [32-34].

In order to increase the performance of those adsorbent materials, activation process such as chemical impregnation, esterification, and carbonization could be conducted as well as combining adsorption to other treatment methods [42]. Several factors affecting the efficiency and effectiveness of the adsorbent to remove oil particle from oily wastewater should be noticed including sorbent dosage, sorbent bed height, sorbent particle size, feed flow rate, initial concentration of oily solution, pH, temperature, and contact time between adsorbent and adsorbate [1].

Table 8. Natural organic-based adsorbent for oil content removal from industrial wastewater

Adsorbent	Oily wastewater derived from industry	Results	Ref.
Modified Barley straw	Canola oil	Adsorption capacity: 44 mg/g, adsorbent dosage: 1.3 g/L, oil removal efficiency: 94%, sorption time: 60 min, pH: 6.0 - 8.0	[35]
Natural wool fibers	Diesel oil	Adsorption capacity: 430 mg/g, adsorbent dosage: 0.04 g/L, oil removal efficiency: 95%, sorption time: 30 min, pH: 8.0	[36]
Sugarcane bagasse	Emulsified machine oil	Adsorption capacity: 680 mg/g, sorption time: 60 min	[37]
Walnut shell	Standard mineral oil	Adsorption capacity: 560 mg/g, sorption time: 60 min	[38]
Walnut shell	vegetable oil	Adsorption capacity: 0.58 g/g, Sorption time: 60 min	[38]
Chitosan (flake)	Palm oil mill	Adsorbent dosage: 2 g/L, sorption time: 30 min, oil removal efficiency: 99%, mixing rate: 100 rpm, pH: 4.0 - 5.0	[39]
Cotton grass fiber	Petroleum Refinery	Adsorption capacity: 1700 mg/g, adsorbent dosage: 1 g/L, oil removal efficiency: 89%, sorption time: 120 min	[40]
Chitosan (powder)	Palm oil mill	Adsorbent dosage: 0.5 g/L, sorption time: 30 min, oil removal efficiency: 99% pH: 4.0 - 5.0	[39]
Palm shell activated carbon	Palm oil mill	Adsorption capacity: 33 mg/g adsorbent dosage: 80 g/L, sorption time: 24 h, oil removal efficiency: 85% pH: 4.5	[41]
Palm shell carbon magnetic composite	Palm oil mill	Adsorption capacity: 100 mg/g, adsorbent dosage: 80 g/L, oil removal efficiency: 90%, sorption time: 24 h,	[41]
Sago bark	Palm oil mill	Adsorbent dosage: 17.5 g/L, sorption time: 30 min, oil removal efficiency: 45%, pH: 4	[42]
Bentonite	Palm oil mill	Adsorbent dosage: 10 g/L, sorption time: 30 min, Oil removal efficiency: 90%, pH: 4.0 - 5.0	[39]
Silkworm cocoon	Vegetable oil	Adsorption capacity: 1500 m g/g, sorption time: 10 min, oil removal efficiency: 54%	[43]
Eucalyptus bark	Petroleum refinery	Adsorbent dosage: 10 g/L, sorption time: 100 min, oil removal efficiency: 91%, pH: 3	[44]

3.4. Coagulation/flocculation

Coagulation and flocculation utilize chemicals to create a rapid-settling aggregate out of finely divided suspension in contaminated water and wastewater leading to elimination of trace organic contaminants. Some stages including destabilization, entrapment, and aggregation or colloids binding processes could create larger or heavier flock particles. As a consequence, those particles could be easily removed by subsequent settling and filtration [45]. The common mechanism of coagulation/flocculation process is illustrated in figure 1 [30].

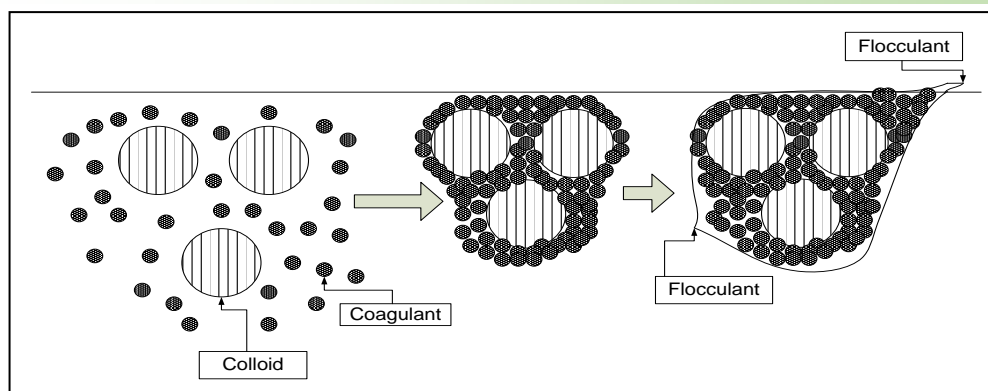


Fig. 1. The mechanism of coagulation/flocculation

There are various coagulants or flocculants available for this purpose such as lime, magnesium, aluminum salts, calcium oxide and aluminum sulphate [46, 47]. Regarding wastewaters containing oil and grease particles, coagulation and flocculation would also be able to break up the chemically stable emulsified oil [2]. This treatment type is usually conducted as pre-treatment step before contaminated water and wastewater entering main treatment system such as membrane filtration or photo-catalysis treatment plant.

3.5. Photocatalytic and oxidation processes

Photocatalytic process is one of the outstanding techniques to enhance the quality of industrial wastewaters as it offers several advantages such as high efficiency, abundant energy sources like solar light, and simple application system, however, costly chemicals usage could be a main concern of this option [48]. This method can be classified into two main processes named homogeneous and heterogeneous processes that can be performed with or without the addition light irradiation. Several metal oxides such as TiO_2 , ZnO , MgO_3 , CeO_2 , ZrO_2 , SnO_2 , WO_3 , $\alpha\text{-FeO}_3$, ZnS , CdS , CdSe , WS_2 , and MgS_2 could be used as photocatalyst agents to succeed the treatment [49].

An investigation on restaurant wastewater containing oil and grease compounds treated using photo-catalytic with TiO_2 addition reported that 10 min irradiation time was sufficient to achieve removal efficiencies of COD, BOD_5 and oil by 50%, 37%, and 86%, respectively [13].

Other oxidation processes named Fenton, Fenton-like, and photo Fenton have also been acknowledged as effective and efficient techniques. Fenton process is based on the use of chemicals known as Fenton's reagents ($\text{Fe}^{2+}/\text{H}_2\text{O}_2$) where the presence of H_2O_2 as an oxidizing agent in wastewater has a positive effect on the degradation rate of the organic pollutants due to more generation of hydroxyl radicals.

3.6. Ozonation process

Ozonation technique relies on the ozone molecule consisting of three negatively charged oxygen atoms to purify wastewater. Ozone could be artificially produced by oxygen generator to remove some pollutant particles including chlorinated hydrocarbons, pesticides, alcohol, ethers, microorganism and aromatic hydrocarbon [50, 51]. A study conducted to treat oily wastewater generated by olive oil manufacturer using hybrid ozone and UV irradiation reported that more than 80% of phenol compounds can be removed while COD concentration could be reduced by 60% [52].

3.7. Biological treatment methods

Biological treatment involving the biological activities of microorganism to treat municipal and industrial wastewater offers some advantages such as being less expensive and non-toxic end products even though this process requires relatively larger plant area, and longer reaction time [53-55]. This is generally applied as primary treatment along with other pretreatment process prior to hydrolyzing oil and grease. In terms of the need of oxygen for them to be active, bacteria used in this process could be aerobic, anaerobic, or both of them. Compared to aerobic, anaerobic treatment tends to generate more biogas and less biomass [56], though anaerobic treatment is still challenging as it could experience disintegration of sludge and limitation on bacteria efficiency due to the overload of oils and fats content [57].

3.8. Integrated treatment methods

It is believed that the application of integrated or multiple methods for treating oily wastewaters will result in better output as well as reduce the rate of machine breakage. Integrated or hybrid wastewater treatment is a prominent solution taken as preferred technology to get the highest quality of treated wastewater [58]. In integrated modes, two or more treatment techniques are applied in sequence. Therefore, some disadvantageous aspects of individual method can be covered by the benefit of other method and vice versa. There are two main parts which are included in an integrated treatment method namely pre-treatment and main treatment stages. In other words, it can include primary and secondary treatment processes [59]. Taking a membrane material as a prevalent example of a method that needs pre-treatment. By reducing the concentration of oil and grease, dissolved and suspended solid contained in the oily wastewater through pre-treatment process, membrane will perform better and longer to produce high quality permeate [22]. Raw wastewater can easily clog and produce fouling on membrane pore and surface leading to fast costly membrane material changing. Therefore, the implementation of some relevant pre-treatment on oily wastewater such as coagulation/flocculation, adsorption or advanced oxidation could elude more severe membrane fouling, reduce chemical cost, and lengthen membrane lifespan [60-66].

Along with other techniques, adsorption has been extensively applied as a pre-treatment stage to reduce targeted pollutant concentration as adsorbent enables those pollutant particles to be trapped in adsorbent's large surface area and abundant micro pores quantities [62, 67]. This has been proved by some experimental investigations conducting the integration of powdered activated carbon and membrane filtration. They reported that adsorption process could reduce cake layer thickness on membrane pores yielding higher permeation flux and pollutant removal efficiency [28, 68]. Advanced oxidation process and coagulation could also be implemented as pre-treatment for oily wastewater derived from various industries including petroleum refinery as they could reduce dramatically any organic compounds and dissolved and suspended solids. [29]. After pre-treatment stage, purer industrial wastewater could enter membrane system leading to much lower fouling rate. Figure 2 illustrates an example of integrated wastewater treatment option involving Fenton and membrane filtration system.

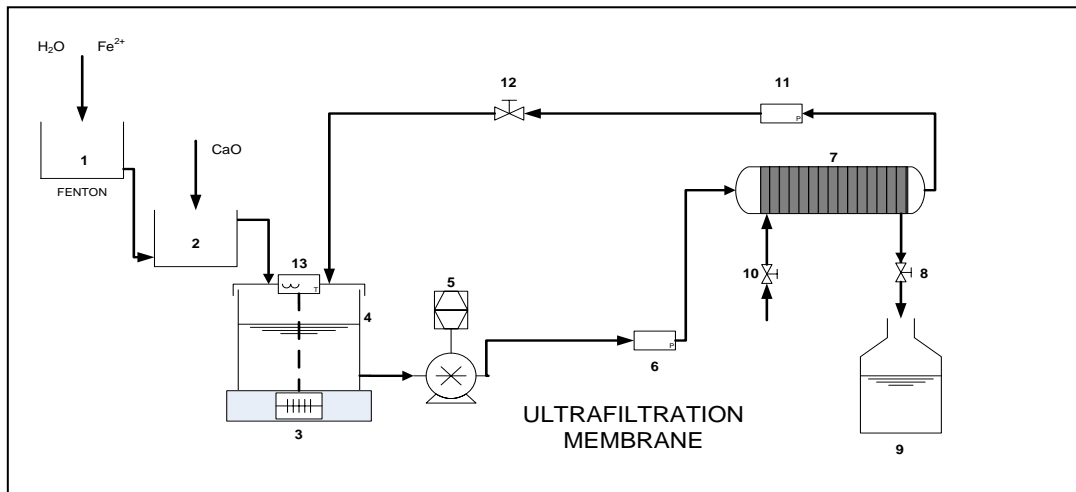


Fig. 2. Integrated membrane system involving pre-treatment process (1: Fenton batch, 2: Coagulation batch, 3: Magnetic stirrer, 4: Feed tank, 5: Digital peristaltic pump, 6: Pressure gauge, 7: Ultrafiltration membrane, 8: Permeate valve, 9: Permeate vessel, 10: Closed valve, 11: Pressure gauge, 12: Retentate valve, 12: Thermometer.

Dual membrane system can also be an alternative to keep membrane performance well. A combination of ultrafiltration and reverse osmosis membrane types for treating oily wastewater, for example, was reported working much better by experiencing only about 7% of permeate flux decline during filtration process [69]. However, it is undeniable that the main weakness of dual membrane treatment system is restricted to the high cost for initial constructing and maintaining works.

4. Conclusion

This review article covers several prominent technologies for treating oily wastewater generated by various industrial sectors including edible oil manufacturers, petroleum refinery, and restaurant industries. As rapid industrialization has generated more amount of wastewater containing oil compounds threatening human health and environmental sustainability, the need of appropriate treatment methods such as membrane filtration, coagulation/flocculation, biological treatment using aerobic or anaerobic bacteria, adsorption, and relevant hybrid methods is unavoidable. These treatment technologies have to be implemented to increase the quality of industrial wastewaters before entering final disposal places. Among others, integrated wastewater treatment methods have been preferred as the best option as they offer better output and safer condition for machine and wastewater treatment plant system. Overall, by studying the existing relevant technology to treat an increasing number of oily wastewater sources, the quality of environment, human lives, and other living creatures will all be benefitted. It also supports the increasing trend of having less waste by doing recycling to support circular economy community as treated water and wastewater can be reused for further usage including irrigation, water process, and even drinking water.

Notation

COD = chemical oxygen demand, mg/L
BOD = biological oxygen demand, mg/L
TOC = total organic carbon, mg/L

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