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Review

Review of Biological Processes in a Membrane Bioreactor (MBR): Effects of Wastewater Characteristics and Operational Parameters on Biodegradation Efficiency When Treating Industrial Oily Wastewater

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Abstract: Oily wastewater is generated from various sources within the petrochemical industry, including extraction, refining and processing, storage, and transportation. Over the years, large volumes of oily wastewater from this industry have made their way into the environment, negatively affecting the environment, human health, and the economy. The raw waters from the petrochemical industry can differ significantly and have complex features, making them difficult to treat. Membrane bioreactors (MBR) are a promising treatment option for complex wastewater; it is a combined physical and biological treatment. The biological component of the MBR is one of the main contributing factors to its success. It is important to know how to control the parameters within the bioreactor to promote the biodegradation of hydrocarbons to improve the treatment efficiency of the MBR. There have been many reviews on the effects of the biological factors of membrane fouling; however, none have discussed the biodegradation process in an MBR and its impact on effluent quality. This review paper investigates the hydrocarbon biodegradation process in an aerobic MBR system by gathering and analyzing the recent academic literature to determine how oily wastewater characteristics and operational parameters affect this process.

Keywords: membrane bioreactor; oily wastewater; bacteria; aerobic; effects on biodegradation

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1. Introduction

Petroleum oil is a valuable strategic resource for which countries compete aggressively. Anthropogenic activities are dependent on oil to meet their energy demand, causing the petrochemical industry to thrive [1,2]. Over the years, large volumes of oily wastewater from petrochemical extraction, refining and processing, storage, and transportation have been discharged into the environment [2,3]. Pollution from oily wastewater manifests itself in many ways, including affecting drinking water and groundwater reserves, endangering aquatic resources, negatively impacting human health, and destroying natural landscapes [4]. Wastewater from various aspects of the petrochemical industry has diverse and complex characteristics, with varying amounts of oil, surfactants, salt, and numerous other chemicals that make it very difficult to treat [5–7]. The oil in wastewater may also be present in different forms: free, dispersed, and emulsified, which are categorized by size. Free oil has droplet sizes bigger than 150 μm, dispersed oil ranges between 20–150 μm, and emulsified oil has droplet sizes smaller than 20 μm [8].

Different treatment strategies are required to remove different components and forms of oil from diverse sources of oily wastewater [7,8]. The traditional methods of treating oily wastewaters use physical, chemical, or biological methods, each having its strengths and disadvantages [9]. In recent years, membrane bioreactors (MBRs), which couple biological and physical treatment, have received more attention. MBRs are easy to operate, have a small footprint, produce high effluent quality by having an enhanced ability to remove contaminants, and generate smaller amounts of sludge than traditional methods [3,6]. The biological element is a key component of a MBR that significantly impacts its effectiveness and efficiency in treating oily wastewater. The microbes found in the activated sludge biodegrade the organic components in raw water before it is filtered through the membrane. Effective treatment of the oily wastewater is dependent on factors that influence microbial activity, such as temperature, pH, the composition of wastewater, the food to micro-organism (F/M) ratio, aeration rate, the mixed liquor suspended solids (MLSS) concentration, sludge retention time (SRT), and hydraulic retention time (HRT), in the bioreactor [6,10,11]. It is vital to know the effects of each parameter and learn to manipulate them in order to achieve optimal conditions within the membrane bioreactor for attaining high-quality effluent with minimal process disruption, such as membrane fouling.

Previous reviews have not concentrated on an MBR system's biological component regarding biodegradation efficiency and effluent quality but rather its contribution to membrane fouling. For example, a few studies reviewed the effects of temperature, pH, salinity, MLSS, F/M ratio, aeration rate, SRT, and HRT on extracellular polymeric substances (EPS) and soluble polymeric substances (SMP) production, which impacts membrane fouling [11–15]. Ouyang and Liu (2009) evaluated transmembrane pressure, MLSS, sludge production rate, specific oxygen uptake, particle size, and membrane fouling when treating domestic wastewaters at different SRTs [16]. Tan et al. (2019) [3], De Temmerman et al. (2014) [17], Di Trapani et al. (2014) [18], and Pendashteh et al. (2011) [19] specifically studied the effect of salt stress on sludge properties and membrane fouling. These studies do not focus on the effects various parameters have on the biomass's metabolic activity. Additionally, none of these studies were specific to oily wastewater. Although there have been many experimental papers that investigate the use of membrane bioreactors to treat oily wastewater and evaluate different aspects of membrane bioreactor performance, there is a lack of summaries of the effects of treatment parameters on bacterial mass in terms of biodegradation capabilities within an MBR [10,20–27]. With the expansion of the application of MBR technology for treating industrial oily wastewater, a comprehensive analysis and review of past academic research on controlling biological activity in an MBR would be valued. It can help in understanding the biodegradation process within an MBR and how it can be optimized.

This review paper compiles the literature from recent research to understand the effects of oily wastewater characteristics and operational parameters on biodegradation within an aerobic membrane bioreactor system. The present review first discusses the sources of oily wastewater from the petrochemical industry to understand its diversity and complexity. It then examines traditional treatment methods, their inadequacies, and the importance of finding new advanced treatment technologies, such as membrane bioreactors, when treating complex wastewaters. A key component of MBR is the biological degradation of organic compounds; therefore, it is crucial to identify the desired type of bacteria and determine how it metabolizes unwanted organic species, in this case, oil. Lastly, it is essential to understand how bacteria react in an MBR and its biodegradation efficiency when treating oily wastewater.

2. Sources of Oily Wastewater

Crude oil is the world's leading non-renewable energy resource, with its demand expected to rise in the coming years [28]. Crude oils are predominantly hydrocarbons, molecules made up of carbon and hydrogen, with hydrogen to carbon ratios ranging between 1.5 to 2. There are over 17,000 organic compounds found in crude oil, which can be classified into four main categories: paraffins, aromatics, asphaltenes, and resins [29,30]. Among them, paraffins are saturated linear chains ranging from a single carbon (methane) to waxes, containing over 40 carbon chains. Aromatic species are benzene rings containing pendant alkyl groups [29,31]. All aromatic compounds contain at least one benzene ring; they are unsaturated rings that react readily due to their lack of hydrogens. Aromatics are extremely recalcitrant due to their high molecular weight, strong molecular bonds, hydrophobicity, and low water solubility [32]. About 15% of crude oil comprises molecules containing heteroatoms, such as oxygen, sulphur, and nitrogen, known as polars, asphaltenes, resins, or NSO [31].

The drilling, extraction, refining, processing, storage, and transportation of crude oil produce lots of oily wastewaters [2,4,28]. Petrochemical wastewater, such as oilfield-produced water, is generated during oil extraction from onshore and offshore wells, which are comprised of high concentrations of artificial surfactants, polymers, radioactive substances, benzenes, phenols, humus, polycyclic aromatic hydrocarbons (PAHs), and emulsified crude oil and are characterized by high COD and low biodegradability [5–7]. Petroleum processing and refinery wastewater can highly differ in composition depending on the operational units for various products. Oil refineries can produce over 2500 refined products that contain different concentrations of ammonia, sulfides, phenols, benzo, and other hydrocarbons [5,6]. Once extracted, crude oil must be carried from extraction wells to oil refineries; after the finished product is obtained, it must be delivered from refineries to distributors. Today, oil tankers are the only option to transport crude oil across oceans worldwide. Between filling and drawing crude oil from and into the oil tankers, the hulls are cleaned with salt water to remove hydrocarbons from the tankers' walls. This operation generates a shipboard industrial effluent known as "slop", distinguished by elevated salt, hydrocarbon, and other organic contaminant concentrations [5,20]. All of these processes associated with the petrochemical industry generate a variety of physically and chemically complex oily wastewaters that need various strategies, including a combination of traditional approaches for effective treatment.

3. Conventional Oily Wastewater Treatment Methods

Conventional oily wastewater treatment methods can be classified into three approaches, chemical, physical, and biological [9].

3.1. Physical Treatment Methods

Gravity separation is a simple and low-cost method that uses the difference in density between oil and water to promote separation. It is most effective at treating low density and viscosity oils due to the greater density difference between the oil and water [32,33]. This process is suitable for separating free and dispersed oil [9]. Gravity separation, however, requires a large area for setup and is not effective at separating emulsified oil [9,34].

Dissolved air floatation (DAF) is a physical treatment system that introduces pressurized air at the base of a basin, and as the fine bubbles rise, it sticks to the oil droplets, carrying oil to the top of the tank [9]. Since oil density is lighter than water, the layer of scum is separated from the water and skimmed off [4]. DAF produces a high-quality effluent that is effective at separating emulsified oil; additionally, it requires a much smaller footprint than gravity separation. One disadvantage of this system is the high operational cost due to the generation of bubbles [4,35,36].

Adsorption is a method where a solid adsorption material with suitable porosity and high surface area is used to absorb medium- to small-sized oil droplets from wastewater [6,9,34]. Conventional adsorption materials, such as zeolite and activated carbon, have a high cost, long adsorption times, and limited adsorption capacity. Currently, research has been focused on developing cheaper new materials, such as foam, biomass, metal-organic framework, chitosan, cotton, sponge, and magnetite nanoparticles to increase adsorption capacity. Although adsorption methods are easy to use and effective, they generate by-products that need further treatment [9,34].

Filtration is another physical treatment option that permits the passage of water through a porous media and traps emulsified or dispersed oil droplets. Examples of such materials are mesh, porous hydrogel, aerogel, textile, and membranes [34]. This method can also recover oil and remove other pollutants from the wastewater. Some disadvantages of this process are the high energy requirement and frequent media cleaning or replacement due to fouling events [37,38].

3.2. Chemical Treatment Method

Coagulation and flocculation is a chemical treatment method where a flocculant is added to the wastewater to neutralize the negative charges of the emulsified or dispersed oil droplets, which helps link particles to form larger flocs [4,9,39]. This process is easy to use and has lower capital and operational cost than DAF and biological technologies [9,34,39,40]. This process's effectiveness relies on flocculant type, dosage, initial oil concentration, and the temperature and pH of the wastewater. Both inorganic and organic flocculants are used for the treatment of oily wastewater. Some commonly applied inorganic flocculants are aluminum sulfate, polymerized ferrous sulfate, and poly-aluminum chloride (PAC), which are cheap and simple to use; however, they display low flocculation efficiency and often require pH adjustment of the wastewater. Organic flocculants, such as polyacrylamide can attain higher flocculation at lower dosages than inorganic flocculants and can be effectively used at wide pH ranges. The downside of organic flocculants is that they present a hazard to the environment and human health as they are challenging to biodegrade [9]. Major disadvantages of the coagulation and flocculation method as a whole are that it produces large quantities of sludge that require secondary treatment and do not work effectively when surfactants are present [4,9,34,39,40].

3.3. Biological Treatment Methods

Biological treatment methods rely on bacterial metabolism to break down hydrocarbons. Conventional activated sludge (CAS) is an aerobic biological treatment that uses suspended microbial floc to treat emulsified or dissolved oil in wastewaters [34,41]. It is cheap and does not require any addition of chemicals during treatment. However, this process requires a large area, long treatment times, and has low treatment capacity [34,38].

In biofilm methods, a biofilm is formed by growing micro-organisms on a filter material or carrier; when the wastewater contacts the biofilm, the micro-organisms metabolize the organic pollutants by using them as nutrients [34]. The support material of the biofilm shelters the micro-organisms from the harsh wastewater conditions, such as high pollutant concentrations and mechanical stress, increasing the survival rate of the immobilized cells and its pollutant biodegradation capabilities. Immobilizing micro-organisms in an appropriate matrix is advantageous for treating heavy oil-polluted wastewaters [41,42]. Although this method is very effective, it has limited operational time due to the formation of multiple cell layers that cause diffusion resistance of substrate and nutrients to the micro-organisms, decreasing its treatment effectiveness [38]. Novel technologies that use biofilms are moving bed bioreactors (MBBR) and sequential batch biofilm reactors (SBBR) [34].

Each treatment method successfully treats certain aspects of the complex oily wastewater generated from the petroleum industry. Therefore, integrating different traditional treatment methods into a single train is the only way to meet discharge or water reuse standards for oily wastewater [9]. Typically, the first step in an integrated system is a physical treatment to remove free and dispersed oil, followed by either a chemical or biological treatment to remove emulsified oil [5]. The standard methods for treating oily wastewater cannot be used as standalone treatments and require a lot of space, have high energy consumption, have long treatment times, and produce secondary pollutants; therefore, it is crucial to find new technologies to overcome these limitations [9,22].

4. Membrane Bioreactor (MBR) Technology

A promising standalone treatment that has proven to be effective at treating oily wastewater generated from the petrochemical industry is a membrane bioreactor. A membrane bioreactor combines biological and physical treatments [14,34,43,44]. It is easy to operate, generates high-quality effluent, and has slower sludge production than CAS [3,6]. The bioreactor component contains activated sludge, which degrades hydrocarbons using microbial metabolism. The bioreactor permits more precise control and management of biodegradation factors including temperature, pH, oxygen, nutrients, and homogenous distribution of hydrophobic contaminants and biomass in the reactor [5]. MBRs allow for a higher mixed liquor suspended solids (MLSS) concentration and sludge retention times (SRT) than CAS, allowing the biomass to develop, adapt, and biodegrade oil more effectively [45,46]. Membranes are semi-permeable barriers through which selectivity between species can be obtained to separate unwanted and wanted particles. They allow the passage of desired species and block the passage of undesirable ones [47]. Microfiltration and ultrafiltration membranes are most effective at treating oily wastewaters [7,9]. MBR technology is capable of removing stably dispersed oil droplets ($<10\ \mu m$) in wastewaters [48,49]. The treatment efficiency of the system can be controlled through operational parameters, such as mixed liquor suspended solids (MLSS), aeration rate, hydraulic retention time (HRT), sludge retention time (SRT), and membrane flux, to achieve optimal treatment conditions that improve membrane performance [7,9,11,14,15,41,50]. Biodegradation is an essential feature of MBR that considerably affects the permeate quality when treating oily wastewater. MBRs are inoculated with activated sludge that contains bacteria that metabolize organic compounds in raw water. Before discussing operational parameters that can control biodegradation efficiency within an MBR, it is essential to understand which bacteria metabolize hydrocarbons and how they degrade them.

5. Biodegradation of Hydrocarbons by Bacteria

Since the discovery of the first hydrocarbon-degrading bacteria, over 175 genera of oil-degrading bacteria have been found. Bacteria from diverse phyla, such as Proteobacteria, Flexibacter-Cytophaga-Bacteroides (CFBs), Actinobacteria, and Cyanobacteria were isolated from different environments, and their effectiveness at metabolizing hydrocarbons was confirmed. Xu et al. (2018) summarize the main petroleum hydrocarbon biodegradation profiles of different bacteria (as shown in Table 1). The biodegradability of oil components usually declines in the sequence of n-alkanes, branched-chain alkanes, branched alkenes, low-molecular-weight n-alkyl aromatics, monoaromatics, cyclic alkanes, PAHs, and asphaltenes [2,51,52]. Alkenes and alkynes are linear unsaturated molecules uncommon in crude oils but are plentiful in refined products such as gasoline [31]. Crude oil and refined oils are complicated and have physical and chemical properties that depend on their constituents and proportion. A slight alteration in constituents can lead to an overall change in physical properties and chemical toxicity [52]. Each strain is able to metabolize a specific type of hydrocarbon [53]. Most bacteria in a genus can only degrade a small range of hydrocarbons with similar structures. For instance, a genus can metabolize alkanes containing different carbon chain lengths, while another can utilize aromatic hydrocarbons with similar characteristics. A single bacterial strain or genus cannot degrade every oil component because of its complicated composition. For example, Alcanivorax metabolizes straight-chain or branched alkanes; Cycloclasticus is known for utilizing PAH as a carbon source [29,30]. Effective biodegradation of various hydrocarbons found in oily wastewaters involves a mixed population of hydrocarbon-metabolizing bacteria with suitable tolerance to environmental changes within a reasonable range for favorable micro-organism activity [29,52].

Table 1. Petroleum hydrocarbon biodegradation profile of bacteria [2].

Petroleum Hydrocarbon Components	Bacterial Species	Main Degradation Profile
Aliphatics	*Dietzia* sp.	n-alkanes (C6-C40)
	Pseudomonas sp.	n-alkanes (C14-C30)
	Oleispira antartica	n-alkanes (C10-C18)
	Rhodococcus ruber	n-alkanes (C13-C17)
	Geobacillus thermodenitrifican	n-alkanes (C15-C36)
	Rhodococcus sp.	Cyclohexane
	Alcanivorax sp.	n-alkanes and branched alkanes
	Gordonia sihwensis	Branched and normal alkanes
Aromatics	*Achromobacter xylosoxidans*	Mono-/polyaromatics
	Aeribacillus pallidus	Mono-/polyaromatics
	Mycobacterium cosmeticum	Monoaromatics
	Pseudomonas aeruginosa	Monoaromatics
	Cycloclasticus	Polyaromatics
	Neptunomonas naphthovoran	Polyaromatics
	Bacillus Licheniformis	Polyaromatics
	Bacillus Mojavensis	Polyaromatics
	Sphingomonas, Sphingobium, and Novosphingobium	Polyaromatics
Resins and asphaltenes	*Pseudomonas* sp.	Resins
	Pseudomonas spp.	Asphaltenes
	Bacillus sp.	Asphaltenes
	Citrobacter sp.	Asphaltenes
	Enterobacter sp.	Asphaltenes
	Staphylococcus sp.	Asphaltenes
	Lysinibacillus sp.	Asphaltenes
	Bacillus sp.	Asphaltenes
	Pseudomonas sp.	Asphaltenes

Before bacteria can start metabolizing hydrocarbons, it needs to be able to access them easily. The proteins and lipids on the microbial cell surface, as well as the biosurfactants that the cell produces, are used to access oil at a wide range of pH levels, temperatures, and salinities [9]. Biosurfactants are vital substances that increase the efficiency with which bacteria absorb petroleum hydrocarbons [2,32]. The bacteria must first attach itself to the oil droplet through pili or flagella. It then secretes biosurfactants of various molecular sizes and chemical natures that emulsify the oil droplet to increase the oil-water surface area and its solubility [30,32,53]. Table 2 shows the different types of biosurfactants that bacteria produce. Bacterial biosurfactant production is natural, non-toxic, and biodegradable, and is a cost-effective technique for assisting in the solubilization of oily wastewater during biodegradation [32,54]. Once the bacteria have solubilized the oil, they can absorb and metabolize the hydrocarbon [55].

Table 2. Biosurfactants produced by hydrocarbon-degrading bacteria.

Bacteria	Biosurfactant	Reference
P. aeruginosa	Rhamnolipids	[51,55]
Rhodococcus sp.	Trehalolipids, Glycolipids	[55,56]
B. licheniformis	Peptide-lipid	[55]
P. fluorescens	Viscosin, Rhanmolopids, Glycolipids, Lipopeptides	[51,55–57]
B. subtilis	Surfactin, Glycolipids, Lipopeptide, Glycopeptide, $^{\varepsilon}$-poly-L-lysine	[51,55–57]
Microbacterium sp.	Carbohydrate-protein-lipid	[55]

A. borkumensis	Glycolipids, Glucose lipids, Trehalose lipids	[58]
P. nautica	Polymeric biosurfactants, lipid-carbohydrate-protein, Glycolipids, Rhamnolipids	[58]
Marinobacter sp.	Carbohydrates: lipids complex, phospholipopeptide	[57]
S. saprophyticus	Glycolipid	[57]
B. circulans	Lipopeptide	[57]
B. mojavensis	Lipopeptide	[57]
B. megaterium	Lipopeptide	[57]
Marinobacter sp.	Carbohydrates: lipids complex, phospholipopeptide	[57]
S. saprophyticus	Glycolipid	[57]
B. circulans	Lipopeptide	[57]
B. mojavensis	Lipopeptide	[57]
B. megaterium	Lipopeptide	[57]
S. lentus	Glycolipid	[57]
E. cloacae	EPS (emulsifier-stabilizing agent in food)	[57]

5.1. Biodegradation of Linear Hydrocarbon Chains

O_2 activates the aerobic alkane degradation pathway. Mono-oxygenases are alkane-activating enzymes that can surmount the low chemical reactivity of hydrocarbons by producing reactive oxygen species. When methane is oxidized, it becomes methanol, then is successively converted into formaldehyde, and lastly, into formic acid. The newly formed formic acid can then be transformed to CO_2 or integrated for the biosynthesis of multi-carbon compounds through the monophosphate or serine pathway, depending on the micro-organism [2,59].

N-alkanes comprising two or more carbon atoms are usually degraded by first oxidizing the terminal methyl group to form a primary alcohol for terminal oxidation. It is then further oxidized to form corresponding aldehydes, and lastly, it is transformed into a fatty acid. The fatty acids are then coupled with CoA and processed by b-oxidation to render acetyl-CoA [2,59]. Acetyl CoA may be used for many other biochemical processes, such as the tricarboxylic acid (TCA) cycle, to produce adenosine triphosphate (ATP). Oxidation of subterminal n-alkanes occurs by converting alkane groups into secondary alcohols and turning them into corresponding ketones. A Baeyer–Villiger mono-oxygenase then oxidizes it to generate an ester. An esterase further hydrolyzes it to alcohol and a fatty acid. Terminal and subterminal oxidation can exist simultaneously in select micro-organisms [2,51,59–61]. The biodegradation pathway of alkanes by aerobic bacteria, as illustrated by Brzeszcz & Kaszycki (2018) [61] is shown in Figure 1.

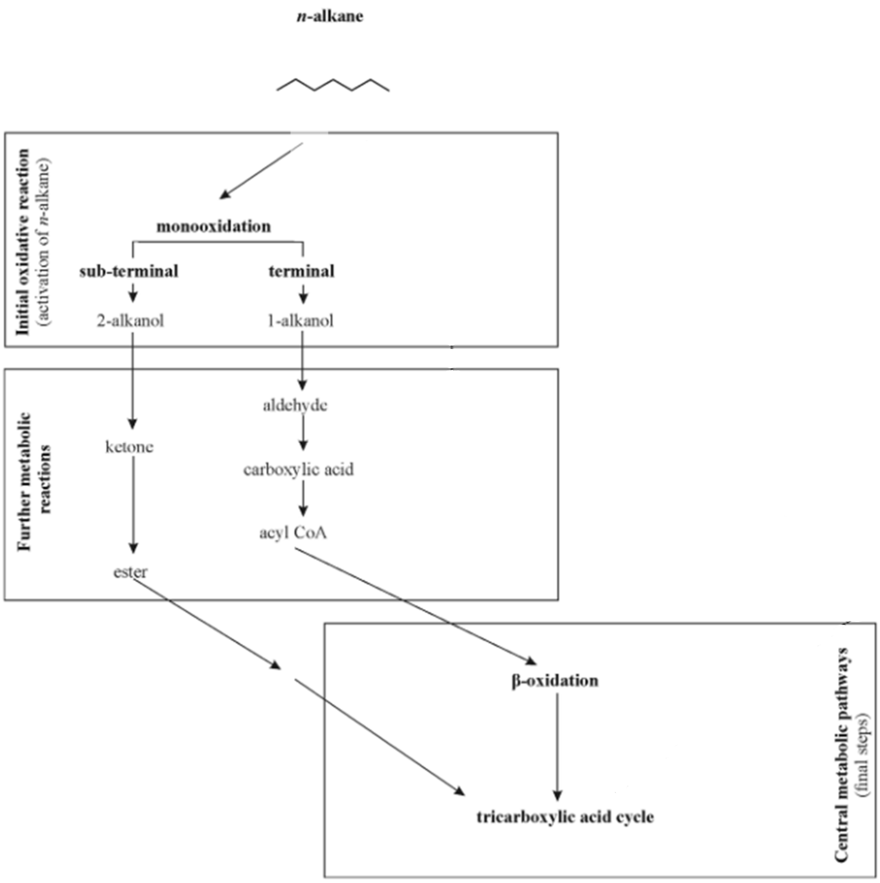

Figure 1. Biodegradation pathways of alkanes by aerobic bacteria "modified figure from [61]".

5.2. Biodegradation of Aromatic Hydrocarbons

PAHs in oil are exceptionally resistant to biodegradation due to the inherent stability of aromatic rings. PAHs are toxic, carcinogenic, and persistent in oil-polluted regions. Although PAHs are difficult to biodegrade, it is not impossible since they are of biological origin. Since they can be found in nature, some micro-organisms exist that can biodegrade them. PAHs are composed of two or more aromatic rings with various branches and aromatic groups [62]. Some well-known bacteria that have the ability to metabolize PAHs are of the genera Arthrobacter, Burkholderia, Mycobacterium, Pseudomonas, Sphingomonas, and Rhodococcus [30].

Naphthenes are the simplest polycyclic aromatic compounds that comprise parent compounds such as cyclopentane, cyclohexane, and decalin, along with their alkylated analogues [31]. They are composed of two fused benzene rings; their chemical formula is $C_{10}H_8$ [32,62]. One of the ways bacteria break down this component is to oxidize naphthalene with mono-oxygenase and dioxygenase attack of the aromatic ring, which yields the intermediate compounds of dihydrodiol. The bacteria utilize the dioxygenase reaction to oxidize naphthalene to D-trans-1,2-dihydroxy-1,2-dihydronaphtalene, then the dehydrogenase enzyme is used to catalyze the previous intermediate to 1,2-dihydroxynaphthalene [2,29,32,63,64]. The dihydroxylated PAH then goes through cleavage by breaking the aromatic ring to create carboxylated compounds, which, if further oxidized by enzymes, are directed to the tricarboxylic acid cycle [51,63,64]. The metabolism of Naphthalene is shown in Figure 2. Bacteria similarly degrade other aromatics. Figure 3 shows the overall biodegradation process of any hydrocarbon by bacteria.

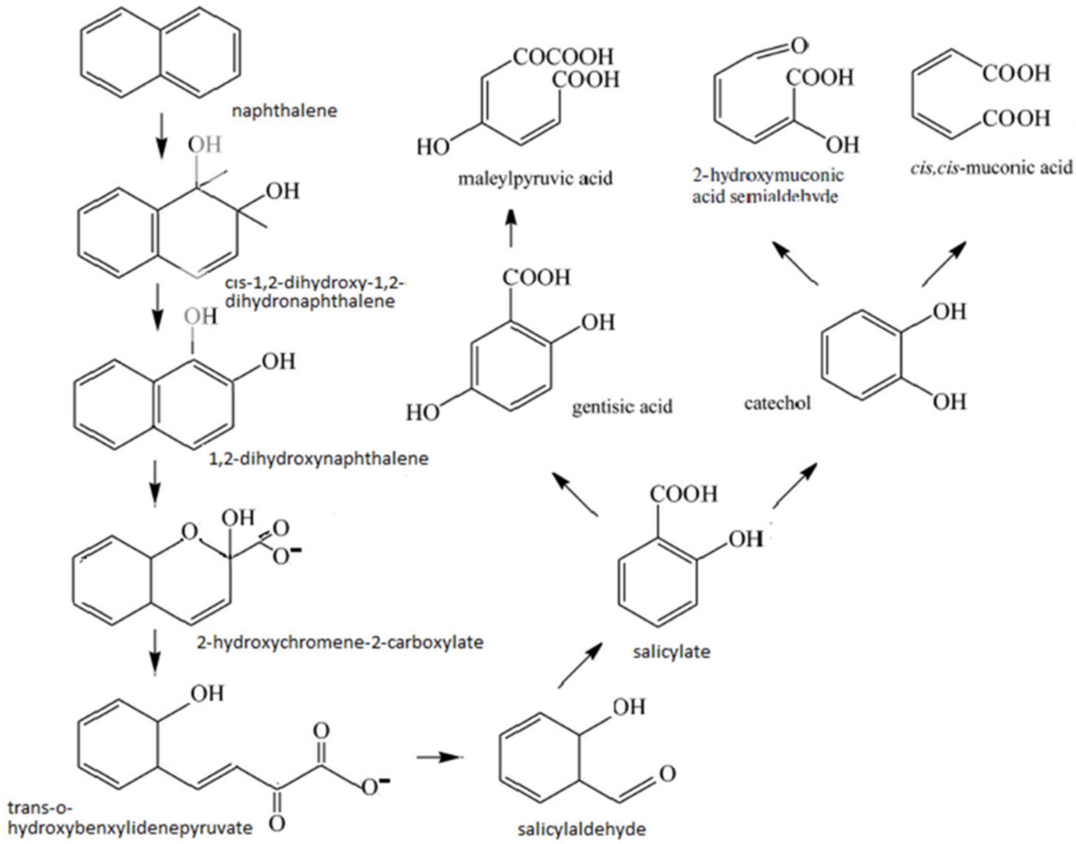

Figure 2. Biodegradation pathway of PAH by aerobic bacteria [63].

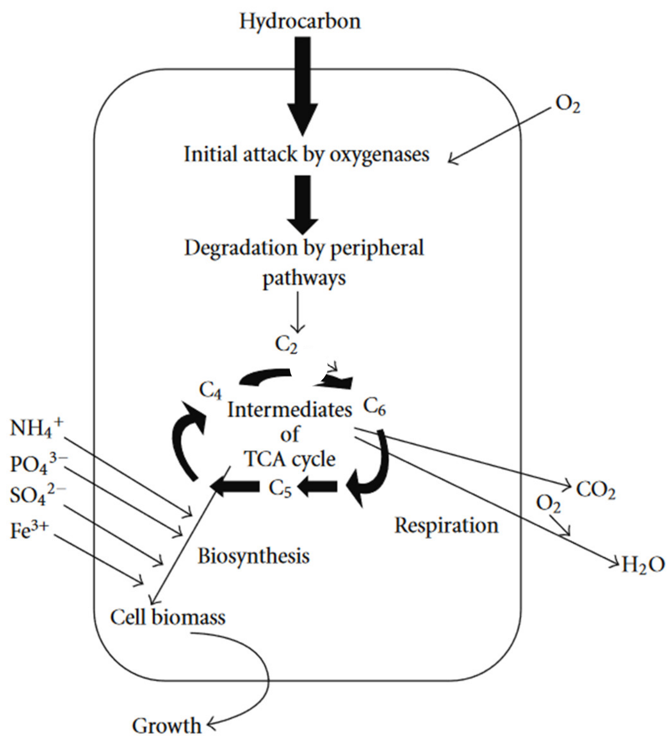

Figure 3. Biodegradation pathway of hydrocarbon by bacteria [51].

6. Effects of Various Parameters on Hydrocarbon Biodegradation within an MBR

Biological processes are a crucial part of MBR; therefore, it is important to understand the effects of wastewater characteristics and the mechanism needed to control microbial activities in the bioreactor [32]. The bacteria must be able to synthesize enzymes that catalyze the reaction of metabolizing hydrocarbons, converting them to simpler or less toxic components [32]. Unlike biodegradation in nature, there are limiting resources within the bioreactor [31]. Microbial activity is contingent on parameters such as wastewater composition, pH, temperature, aeration, F/M ratio, hydraulic retention time, and sludge retention time [41,65].

6.1. Effects of Temperature

Temperature affects the physical nature and chemical composition of hydrocarbons. It determines the hydrocarbons that remain after evaporation and the oil surface area availability for microbial access for biodegradation. At low temperatures, the oil's viscosity increases, reducing the volatilization of toxic short-chain alkanes and the solubility of oil [45,52,60,66]. The rate of biodegradation decreases as well with decreasing temperatures due to the reduced rate of bacterial enzymatic activity [66]. Higher temperatures decrease the viscosity of hydrocarbons and increase the volatilization of BTEX components, solubility of oil and the rate of microbial metabolism [11,14,45,51,66]. The optimal biodegradation temperature of hydrocarbons by bacteria is within the range of 30 to 40 °C; higher temperatures increase the toxicity of hydrocarbons to the bacteria [60,66].

Alsalhy et al. (2016) [67] explored the effects of feed water preheating times (i.e., 15, 30, and 45 min) at a temperature of 45 °C on oil removal in oil refinery wastewater using an MBR. The activated sludge concentration for this set of experiments was characterized by 1000 mg/L MLSS. The oil concentration decreased by 95% with three days of hydraulic retention time when using preheating time of 15 min. The system was capable of removing 100% of oil when using preheating times of 30 and 45 min. Increasing the temperature to 55 °C increased oil removal rapidity. In addition to oil removal, Alsalhy et al. [67] measured the effects of preheating on COD removal. The COD concentration was reduced by 50%, 58%, and 64% when using preheating times of 15, 30, and 45 min, respectively. Increasing the temperature to 55 °C considerably increased COD removal efficiency. The COD removals increased to 52%, 63%, and 71% for preheating times of 15, 30, and 45 min, respectively. The increase in temperature and preheating time removed the volatile substances from the wastewater and promoted the biodegradation of hydrocarbons by making them more accessible to bacteria. A similar study by Al-Malack et al. (2007) [68] confirmed the results.

6.2. Effects of pH

The pH of wastewater affects the biodegradation of hydrocarbons; it impacts processes such as cell membrane transport, catalytic reaction balance, and enzyme activities. Most heterotrophic bacteria favor neutral to alkaline pH in the range of 7.2–8.5 [45,60]. Previous studies have found that the microbial mineralization of naphthalenes and octadecanes can occur at a pH of 6.5. Phenantherenes were biodegraded in liquid media effectively at a pH range of 6.5–7.0 by bacteria such as Burkholderia cocovenenas. Pseudomonas aeruginosa was capable of biodegradation of crude oil in water up to a pH of 8.0 [60].

6.3. Effects of Salinity

Salinity is another factor affecting micro-organisms in an MBR in terms of growth and floc rheology. The enzymatic activity of micro-organisms can be severely inhibited due to toxic effects. It can also impact the structure of the microbial consortia within the bioreactor [17,22,60,69–71]. A sharp increase in salinity results in osmotic pressure on the cell membrane, causing dehydration and eventual plasmolysis, consequently decreasing sludge settleability and bioflocculation [72,73]. The toxic effects of salinity can be overcome by gradually acclimating the bacteria to the high saline conditions [20,21,23,71,74–76].

In recent years, many studies have considered using a microbial consortium that can degrade hydrocarbons and tolerate saline environments to optimize the treatment process when treating saline oily wastewaters [77]. Halophilic and halotolerant marine micro-organisms have been used to inoculate MBRs to improve biodiversity and enhance the efficacy of biodegradation to treat saline oily wastewater [22,78–80].

6.4. Effects of Aeration

Oxygen supply is critical for aerobic degradation as it functions as a terminal electron acceptor in the hydrocarbon metabolism process, making aeration an essential parameter to consider in MBR treatment of wastewater, as it is directly related to DO concentration within the bioreactor [14,32,45,52,60]. It is understood that the mass of oxygen needed to metabolize a hydrocarbon load is approximately 0.3 g of oxygen per gram of oil oxidized [52].

Biomass features, such as SMP and EPS substantially impact the organics removal rate due to their effect on oxygen transport. SMP is soluble and hence exists in the liquid phase, whereas EPS is attached to cells and thus exists in the solid phase. The quantity of EPS varies with changes in the microbial state and bioreactor operation conditions. The oxygen must first permeate the liquid layer surrounding the flocs and then diffuse through the floc matrix (EPS) to reach the active spots on the bacterial cell membrane. Over-aeration can result in poor sludge properties, such as weak floc structure and a low sludge volume index (SVI), causing dispersion of bacteria in the MBR that leads to poor biodegradation [14,45]. On the other hand, low aeration can create anaerobic conditions through excess SMP and EPS production [13,69,81,82]. Thus, it is crucial to find the optimal aeration rate to provide micro-organisms with enough oxygen to perform metabolic processes and at the same time not disturb floc formation.

Additionally, aeration generates unsteady-state shear at the membrane surface via turbulent eddies, fiber oscillations, particle scouring, and recirculation of content in the bioreactor, which contribute to a reduction of cake layer formation on the membrane surface, consequently reducing membrane fouling [14,45,83]. If aeration is inadequate, the membranes are more susceptible to clogging or blocking by bacterial EPS and SMP production, which causes uneven distribution of flow, disrupting permeate production by obstructing permeate flow through the membrane [83].

6.5. Effects of Nutrients

The development of heterotrophic bacteria relies on nutritional elements and an electron acceptor, such as oxygen, for biodegradation with aerobic bacteria [32,51,52]. The lack of any of these elements hinders the growth and metabolism of the micro-organism. Bacteria responsible for degrading hydrocarbon require a fixed nitrogen source such as NH3, NO3-, NO2- (inorganic), and some organic nitrogen sources. Phosphorus is another critical nutrient for the microbial population as it is utilized to synthesize adenosine triphosphate (ATP), nucleic acids, and cell membrane components [32,51].

To function correctly, the activated sludge in MBR requires a balance between the food entering the bioreactor and the bacteria in the bioreactor. A high F/M ratio indicates that there is more food than micro-organisms available to consume that food. When the F/M ratio is high in bioreactors, the bacteria are active and proliferate quickly, but they are also more distributed. Dispersion provides an environment in which the bacteria does not form an adequate, big, dense floc and, as a result, frequently leads to poor settling [60,84]. A low F/M ratio indicates that there are numerous microbes but a limited food supply. When food is scarce, bacteria begin to produce a thicker slime layer, lose their motility, and cluster together to form a dense floc that settles readily [60]. At a high sludge age and MLSS concentration, the MBR systems require fewer nutrients due to the decline in excess sludge production [27,45]. The micro-organisms in the system reach an equilibrium state where the amount of energy provided (food) equals the microbial maintenance demand. The maintenance of biomass is preferred over the production of additional biomass. When the F/M ratio is lowered, the biomass production is reduced, and the food is used for maintenance [12,45,85]. The low nutrient demand for biomass to treat wastewater effectively is an advantage of MBR systems over CAS since the lack of nutrients in industrial wastewaters is common. In comparison, the nutrient requirement for oil biodegradation in a CAS treatment method amounts to 120 g nitrogen and 20 g phosphorus for every kg of oil, whereas MBR only needs 6.7 g nitrogen and 0.8 g phosphorus as nutrients to biodegrade 1 kg of oil [14,27].

6.6. Effects of MLSS

MLSS concentration influences biomass growth; generally, the higher the MLSS concentration, the higher the biomass in a system [12,67]. A high level of MLSS concentration decreases the sludge loading rate, enhances the treatment efficacy, and increases the viscosity of the mixed liquor [86]. The mixed liquor volatile suspended solids (MLVSS) to MLSS (MLVSS/MLSS) ratio is utilized as a measure of the quantity of viable sludge in MBRs [45].

Alsalhy et al. (2016) [67] studied oil refinery wastewater treatment using a membrane bioreactor (MBR). Two different MLSS concentrations (500 and 1000 mg/L) were tested for COD, BOD, and oil removal within 5 days at a constant temperature of 25 °C and HRT of 3 days. The COD, BOD, and oil concentrations of the system reduced from 235 mg/L to 157 mg/L, 46 mg/L to 31 mg/L, and 14 mg/L to 2.3 mg/L when using an MLSS concentration of 500 mg/L, respectively. When using an MLSS concentration of 1000 mg/L, COD, BOD, and oil decreased from 235 mg/L to 122 mg/L, 46 mg/L to 28 mg/L, and 14 mg/L to 1.3 mg/L, respectively. The findings of this study show that with an increase in the MLSS concentration, the removal of COD, BOD, and oil also increases; this can be attributed to the higher biodegradation of hydrocarbons into organic components.

In another study by Capodici et al. (2017) [21], inoculated an MBR unit with a biomass concentration of 4 g/L TSS. Until Day 54, a reduction in suspended biomass was observed. This result can be attributed to the stress effect exerted by the hydrocarbons on the unacclimated biomass. Thus, to maintain biomass activity towards the toxic organic substance, from Day 54, sodium acetate was added to the influent water at a concentration of 500 mg/L. After adding the nutrients, the suspended biomass growth increased up to 7 g/L TSS, indicating favorable acclimation and an increase in biomass activity. The experiments showed that the MBR system was effective at removing COD from oily wastewater; the MBR as a whole achieved a COD removal efficiency of 88%, with the biological component contributing to 70% of the total removal efficiency. The researchers also evaluated the removal efficiency of hydrocarbons and found that the system achieved a TPH removal efficiency of 92%, with a permeate TPH concentration below 5 ppm.

Similarly, Di Bella et al. (2015) [24] evaluated the performance of an MBR system for the treatment of shipboard slops. This study assessed hydrocarbon degradation efficiency and biomass activity under salinity variation. The MBR unit was inoculated with 3.5 g/L TSS of activated sludge from a municipal wastewater plant. Synthetic wastewater with a gradual increase in salinity was fed to the MBR system for 60 days (Phase I); after that, the unit was fed with a mixture of synthetic wastewater and shipboard slops. The percentage of shipboard slops in the wastewater was gradually increased to 50% within 30 days (Phase II). The activated sludge in the system was retained during the 30 days it was fed with increasing slop concentration, resulting in the MLSS concentration of the MBR to increase to 8 g/L TSS. This ensured the biomass acclimation to salinity and hydrocarbons. The COD removal efficiency of the MBR was reported to be approximately between 57–96%, to which organic compounds' biomass degradation contributed approximately 55–90% in Phase I. The MBR achieved a COD removal efficiency of 65–97%, to which 46–85% of the COD removal was attributed to biodegradation in Phase II. The MBR unit attained a 47.5% TPH removal efficiency when slop concentration increased to 50%. These studies show that biomass acclimation to salinity and toxic hydrocarbons is necessary for effective oily wastewater treatment. It can also be seen that biodegradation is a major component in the MBR system, accounting for the majority of the treatment efficiency.

The need for a biomass acclimation period indicates that there is a change in biomass composition that occurs with a changing environmental composition [20,54,87]. In the case of oily wastewater, the abundance of hydrocarbon-degrading bacteria increases while micro-organisms that cannot adapt are gradually eliminated. Likewise, halotolerant and halophilic bacteria in saline wastewater increase while the non-halotolerant and non-halophilic bacteria die off [63]. Additionally, the diversity of bacteria in activated sludge changes with treatment time [63,86]. Different carbon sources ensue changes in equilibrium between bacterial strains in a consortium. When a particular type of hydrocarbon is in contact with the bacteria, the strain that is capable of metabolizing it becomes dominant in the consortia; this phenomenon is called microbial succession [2,54,87–90].

6.7. Effects of SRT

One of the benefits of MBRs is the high sludge age attained over a long SRT compared to CAS. Long SRTs are unattainable in conventional treatment methods because of the inadequate settling capacity of sludge at high concentrations and extraction of suspended particles with the effluent. Typically, high MLSS concentrations due to high sludge age in the MBR allow wastewaters to be treated effectively at long SRTs, which minimize biomass yield and decrease sludge production [45,46]. High sludge age achieved through a longer SRT permits the retention of particulate, colloidal, and higher-weight organics, which provides maximum opportunity for bacteria to metabolize organic compounds and allows for acclimation of microbes to the biodegradable compounds. As a result, biomass adapts to wastewater without being limited to fast-growing and floc-forming microbes [27,45,46,91]. The slow sludge production removes the concern for changing biomass settling characteristics (i.e., filamentous bacterial growth, bacteria dispersion, and floc densification) that disrupt treatment efficiency by worsening sludge filterability and contribute to membrane fouling [12,45,46]. SRT should be chosen to prevent both the negative impacts of accumulating non-biodegradable chemicals caused by low sludge discharge and excessive sludge generation at low sludge ages. Low sludge age under a low SRT can cause foaming and sludge bulking [45].

Kose et al. (2012) [26] studied the effects of SRT on MBR treatment efficiencies of brackish oil and oil field-produced water. The MBR was set up as a continuous flow submerged MBR system and was operated at room temperature for 297 days. Two different SRTs, 30 days and infinite days, were used. The MBR was inoculated with sludge acquired from laboratory scale MBR leachate. This study found that the COD removal efficiency increased from 80% to 85% due to the higher biomass concentration at a higher SRT. With an increase in sludge age, the oil and grease removal efficiency increased from 60% to 85%. At an SRT of infinity, over 99% of TPH was rejected. Various other studies confirmed that longer SRT under-steady state conditions result in better COD and oil removal efficiencies [10,11,14,19,23,76].

6.8. Effects of HRT

HRT is the amount of time that the wastewater remains in the MBR. Generally, the longer the HRT, the longer the contact time between the bacteria and the biodegradable organic compounds in the wastewater. As a consequence, the removal efficiency of the organic compound and COD increase [5,19,91,92]. It is critical to emphasize that reactor performance can only be obtained when MBRs are operated under steady-state settings. The achievement of steady-state depends not only on the length of the operating period but also on the sludge's adaptation to the wastewater components [91].

Razavi and Miri (2015) [10] explored the effects of various HRTs on treating real petroleum refinery wastewater. MLSS concentration in this system was kept between 3–6.6

g/L. The three different HRTs tested were 36 h, 30 h, and 25 h. It was found that reducing HRT from 36 h to 30 h and 25 h yielded COD removal efficiencies of 81.08%, 78.92%, and 78.92%, respectively. BOD removal efficiencies were 86.1%, 87.6%, and 89%, respectively, with increasing HRTs. The findings correspond with similar studies that found that complex oily industrial wastewaters need longer hydraulic retention times for effective treatment once biomass is acclimated to the harsh environment [14,19,92–94].

7. Conclusions

This paper comprehensively reviews the biological processes within an aerobic MBR, since it is a key component that contributes to an MBR's success in treating complex oily wastewaters; the review delved into identifying desired bacteria that can utilize oil as an energy source and the metabolic processes of degrading different types of hydrocarbons. The evaluation was focused on determining the effects of oily wastewater characteristics and operational parameters on biodegradation efficiency in an aerobic MBR system. This review provides an in-depth analysis of factors that control the biodegradation process in an MBR and can be used as a guide to optimize the treatment system.

MBRs are very robust systems capable of handling oily wastewater from various sources within the petrochemical industry. There are vital factors that affect the biological component of MBRs to consider when treating this type of wastewater. The first are raw water characteristics, such as composition, pH, and, temperature, which affect the biodegradation process. With elevated levels of hydrocarbons or salinity, an acclimation period is needed to develop an appropriate bacterial consortium in the sludge. When treating saline oily wastewater, if MBR is inoculated with non-halotolerant bacteria, it is essential to acclimate them beforehand since the salt shock causes the dehydration and plasmolysis of bacteria, quickly killing the biomass and producing excess EPS and SMP, resulting in poor biodegradation and membrane fouling. In recent years halophilic and halotolerant bacteria that are known to degrade various hydrocarbons have been used to inoculate MBRs, which has proven effective, thereby eliminating the need for or reducing the duration of biomass acclimation. Then, there are operational parameters that can be controlled, including MLSS concentration; typically, the greater the MLSS, the lower the organic loading rate, up to a certain point, and the better the treatment efficiency. Adequate aeration is crucial for providing the bacteria with enough oxygen to perform metabolic processes. A low F/M ratio is optimal to provide the bacteria with sufficient nutrients to maintain the biomass and aid their metabolic processes without promoting excess sludge production. SRT is another critical parameter; it should be chosen carefully to suppress the accumulation of non-biodegradable components due to low sludge discharge and excess sludge production at a low sludge age, and to promote a high sludge age through long SRTs, allowing for biomass acclimation and efficient treatment. Lastly, longer HRTs are recommended for treating oily wastewater to allow biomass enough contact time with hydrocarbons to metabolize them effectively. MBRs are very successful technologies for treating industrial oily wastewaters, a possible way to improve the treatment efficiency in terms of biodegradation, future studies can explore the use of genetically modified microbial consortia that can target specific compounds in the raw water.

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