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Review

Review of Hollow Fiber (HF) Membrane Filtration Technology for the Treatment of Oily Wastewater: Applications and Challenges

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Abstract: Oily wastewater has been recognized as a threat to the environment due to its hazardous nature and it can negatively affect the ecosystem, and threaten wildlife and human health. Physical, chemical, and biological technologies demonstrated a mixed performance in oily wastewater treatment, and, therefore, a proper treatment technology for oily wastewater needs to be addressed. Membrane filtration using a hollow fiber (HF) membrane is a promising alternative to remove emulsified oil from oily wastewater. This review discusses different sources of oily wastewater, various treatment methods, and membrane technology. The assessment has been focused on the parameters affecting HF membrane performance and applications of HF membrane-based technology to treat oily wastewater. This review paper reveals that HF membrane filtration systems have been previously used for the treatment of oily wastewater in bench-scale studies and few pilot-scale applications, which proved to be favorable in the treatment of recalcitrant wastewater containing oil and high salinity. Limitations associated with membrane fouling and the reduction of membrane permeability and membrane lifespan can be tackled and alleviated through modifying membrane chemistry and adjusting operational parameters. The compilation of studies showed that a low food/microorganism (F/M) ratio, long solid retention time (SRT) with high sludge age, long hydraulic retention time (HRT), and moderate aeration were the preferred operational parameters when treating oily wastewater. Based on this review, future studies should focus on optimizing the hydrodynamic conditions of the HF system, the commercialization of modified HF membranes, and the utilization of green technology in HF membrane construction to broaden HF membrane technology applications.

Keywords: membrane technology; hollow fiber membrane; oily wastewater treatment; physical separation; membrane bioreactor

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1. Introduction

In the past two decades, different industrial sectors have generated large volumes of oily wastewater and discharged it into the environment [1–4]. Oily wastewater is characterized by high biochemical oxygen demand (BOD), total suspended solids (TSS), chemical oxygen demand (COD), ammonia, sulphides, total organic carbon (TOC), and total petroleum hydrocarbon (TPH), with their concentration varying depending on the operations and products from the manufacturing industries [5]. It also contains many toxic compounds, such as volatile organic compounds (VOCs) (e.g., benzene, toluene, ethylbenzene, and xylene (BTEX)), polycyclic aromatic hydrocarbons (PAHs), phenols, and heavy metals [4,6–8]. When oily wastewater is discharged into water bodies, land, and/or sewer lines without adequate treatment, it negatively affects drinking water and groundwater sources, wildlife, and human health, and also causes atmospheric pollution [9,10]. The global increase in the discharge of oily wastewater, and strict environmental regulations for effluent discharge, necessitate the need for effective and adequate oily wastewater treatment [5]. Previously, a variety of treatments, including physical, chemical, and biological methods have been widely used to treat oily wastewater and mitigate its environmental impacts [11]. However, physical methods require large space for installation, high energy for dispersing coagulants, long periods of time to separate oil using gravitational force, and a low treatment efficiency [3,6,9,12]. Chemical methods consume toxic compounds, have high chemical costs, generate secondary pollutants, and are not suitable for the environment [3,6,9,13]. The efficiency of biological methods is affected by changes in environmental factors such as the level of oxygen, temperature, and variation of feed composition. Oil compounds are highly toxic and have poor nutrients for microorganisms; and several compounds such as saturates, aromatic, asphaltenes, and resins cannot be decomposed efficiently using this method [13,14]. Therefore, the focus of researchers has shifted to emerging and advanced treatment technologies for sustainable, cost-effective, eco-friendly, and efficient treatment of oily wastewater [5].

Among all of the advanced methods, membrane-based technology is a promising method for oily wastewater treatment due to its capability in separating oil droplets smaller than 20 μm [15–17]. The membrane filtration technology has benefits such as high effluent quality, low requirement for chemical additives, low generation of sludge, and a small footprint [8,18,19]. Additionally, membrane technology has a low energy cost compared to conventional and other advanced technologies. Membrane filtration treatment does not have any moving parts, has the possibility of using intermittent aeration, and has low chemical substance usage, which contributes to its low energy requirement [20–22]. Hollow fiber (HF) is the desired membrane configuration for oily wastewater treatment as it has the highest surface area per unit volume, is mechanically self-supported, and can stimulate the movement of hollow fibers through air scouring, and backwashing, which helps mitigate membrane fouling [23,24]. This technology can either be used as a physical separation approach or integrated with a biological component (i.e., membrane bioreactor (MBR)) for treating oily wastewater [18,25]. The application of HF membranes for oily wastewater treatments is contingent on various factors such as membrane material, features, and operational factors such as flux, aeration flow rate, and activated sludge characteristics [26,27]. There needs to be a delicate balance between operational parameters to attain optimal membrane permeability, and, consequently, high effluent quality [28].

Although membrane-based technologies are dependable treatment systems, membrane fouling is a notable problem impeding widespread and large-scale applications of this technology [2,29]. Membrane fouling increases transmembrane pressure (TMP), which, in turn, reduces permeate production and decreases membrane lifespan. Consequently, frequent membrane cleaning and replacement are required, therefore, increasing operational costs [2,28]. Thus, further research is required for developing new methods for enhancing membrane performance.

The objective of this review paper is to investigate the application of HF membrane-based technology and identify its challenges in order to improve its performance in treating oily wastewater. The first section of this paper discusses the sources of oily wastewater and its treatment methods. The assessment then focuses on membrane-based technology, HF membrane characteristics, important parameters affecting its performance, and HF membrane applications in oily wastewater treatment. The paper explores recommendations to improve the treatment of oily wastewater using an HF membrane filtration system.

2. Oily Wastewater and Treatment Methods

2.1. Oily Wastewater Sources

Different industrial sources generate an immense volume of oily wastewater which adversely affects the environment, wildlife, and human health due to the existence of

BTEX, phenols, and total acid and grease with quantities ranging between 0.73–24.1 mg/L, 0.001–10,000 mg/L, and 2–560 mg/L, respectively [30].

Food, metallurgical, petroleum, and transportation industries are among the significant sources of oily wastewater [4,31].

In the food industry, palm, soybean, olive, cottonseed, and sunflower are major sources of extracting edible oil. The global oil market is predicted to increase from 83.4 billion US$ (2015) to 130.3 billion US$ by 2024 with a compound annual growth rate (CAGR) of 5.1% [32]. Palm oil is the most common edible oil in the world with 37% of the total vegetable oil production [33]. During oil processing, a high volume of oily wastewater is generated (e.g., during the production of palm oil, 0.5–0.75 tons of palm oil mill effluent per one ton of fresh fruits were generated) [34]. This type of oily wastewater has a high concentration of COD, BOD, total dissolved solids (TDS), TSS, oil and grease, fats, phosphate, and sulphate [32].

Another significant source of oily wastewater is generated from the lubricating and cooling of metal pieces in machinery [35]. They are spilt into two categories, one is oil-based metalworking fluids (MWFs) and the other is water-based MWFs [36]. The waste generated as a result of using these MWFs is known as spent cutting oil [37]. On a global scale, more than 2,000,000 m^3 of MWFs are used annually, however, the volume of wastewater can be ten times higher because of the dilution of MWFs [36]. MWF is comprised of heavy metals, acids/alkalines, radioactive metals, phenols, PAHs, hydrazines and imine-carbohydrazides, and thiophenes as well as hydrocarbons containing BTEX which are persistent in the environment [38].

Oily wastewater is generated from every step of the petroleum industry including exploration, drilling, processing, and storage, with the wastewater produced from these processes referred to as produced water [4]. Among them, oil drilling and oil refinery processes produce the largest amount of oily wastewater [39]. For instance, the total amount of produced water globally generated from the petroleum industry is 39.746×10^9 L/d [9]. The characteristics of the produced water are varied depending on the location of the refinery and oil well [4,31]. Petroleum wastewater contains a complicated mixture of oil, water, and suspended and dissolved solids. PAHs, phenols, heavy metals, and radioactive elements are among the most concerning components [16,40–42]. Marine transport is a major source of oil pollution; the emergence of the worldwide shipping industry increased illicit discharge of bilge and ballast water and oil spill disasters [11]. For instance, the International Tanker Owners Pollution Federation (ITOPF) recorded the largest volume of oil spilled in the past 24 years (i.e., SANCHI, off the coast of China) occurred in 2018, releasing ~98,000 tonnes of oil [43]. In addition, the oil spill volume for 2021 is the second highest estimate (i.e., ~82,000 tonnes) in the last ten years globally as a result of six oil spill incidents [43]. Harmful compounds that are released in the water as a result of a spill include hydrocarbons, hazardous metal ions, detergents, surfactants, and petroleum products, such as crude oil, diesel oil, gasoline, lubricant, and kerosene [11].

2.2. Treatment Methods

Different types of treatment have been used to separate oil from oily wastewater including physical, chemical, and biological methods [44,45]. A suitable treatment technology is selected based on the source of wastewater, wastewater characteristics, operational cost, and the end-use of effluent [4,5].

2.2.1. Physical Methods

Physical treatment methods include gravity separation, dissolved air floatation (DAF), coagulation, and membrane separation which are used to treat oily wastewater [11,46–48]. The gravity separation method is operated based on the difference in densities of hydrocarbons and water phases, this method is suitable for removing suspended solids (SS) and free oil (i.e., oil–water mixture with droplet diameters >150 μm) [49,50]. Gravity separation is usually used as a pre-treatment or primary treatment; since this method is incapable of separating dispersed oil (i.e., oil droplet size ranging between 20 and 150 μm) or emulsified oil (i.e., droplet size of smaller than 20 μm) [51]. In this approach, gravity as the driving force is very slow, incomplete, and time-consuming in many situations [51].

Compared to gravity separation, DAF is a faster technology and has a smaller footprint. DAF can effectively separate dispersed and emulsified oils, using gas bubbles that are introduced into the oil-containing liquid which sticks to the oil droplets. The gas bubbles quickly rise to the surface demulsifying the solution; the oil layer on top is then removed [52]. Previous studies showed that increasing oil concentration and flow rate decreased the oil removal efficiency in DAF. Increasing the flow rate into the DAF tank can cause the oil droplets to flow straight to the effluent without having much time to attach to the air bubbles and float to the surface [47,53]. Increasing the pH range decreases the oil removal efficiency since it increases the repulsion between the oil and water surface and hinders oil droplets from attaching to the air bubbles in the clarifier due to the absorption of $(OH)^-$ ions at the oil–water interface [53,54]. Increasing the temperature of the oil-in-water mixture decreases the viscosity and the surface tensions which increases the separation rate of oil and water [55,56]. Due to the continuous generation of air bubbles, DAF technology has the disadvantage of high energy consumption [6].

Another technique, coagulation, is often used prior to DAF [57,58]; the coagulation mechanism is based on using coagulants to destabilize the colloids through the neutralization of the repulsive forces between the fine colloids [11,59]. This method is capable of removing emulsified and dispersed oils in addition to some difficult to biodegrade organic polymers [60]. Previous studies used different types of coagulants such as poly-zinc silicate, polyaluminum chloride, polyferric sulphate, polyferricsilicate sulphate, and chitosan to remove oil efficiently [61–64]. Parameters such as molecular weight, dosage, and charge density of coagulants determine the coagulation efficiency [65]. The coagulants with a low molecular weight capable of neutralizing the negative charge on the oil droplet have been commonly used [66]. Studies showed that a low coagulant dosage is not adequate to destabilize all of the colloidal particles and increasing the coagulant dosage increases the oil removal efficiency. However, when the dose of coagulants saturates the solution, there is a decreasing trend in the oil removal efficiency [67]. Coagulants with higher charge density demonstrated good results in oily wastewater treatment, for example, polyaluminum chloride with a high charge density showed an excellent result in oily wastewater treatment because the flocs were large and dense during the coagulation process [68]. Although coagulation is an effective process, it has some drawbacks such as requiring a large amount of coagulants that are hazardous to the environment, causing corrosion problems when pH is reduced, and the generation of sludge that needs further treatment [60].

The membrane filtration system overcomes the weaknesses of other methods in terms of a prolonged process, large-space requirement, high amount of waste generation, and low treatment efficiency [3,6,16]. The membrane technology utilized for oily wastewater treatment is driven by pressure. The membrane pore size acts as a selective barrier allowing the smaller particles to pass through, while the larger-sized oil particles are blocked and retained in the feed solution [69,70]. This method is an excellent approach for the treatment of oily wastewater, particularly for effluents containing emulsified oil with minimal density difference in comparison with water [16,71,72]. The summary of physical treatment methods in terms of advantages and disadvantages is shown in Table 1.

2.2.2. Chemical Methods

Advanced oxidation process (AOP), adsorption, and demulsification are categorized as chemical treatments [77–83]. The AOP is described as the method that relies on the production of hydroxyl free radicals, which has great electrochemical oxidant power and strong oxidizing potential. Their excellent oxidizing potential allows them to easily degrade organic compounds (i.e., oil) converting them to H_2O, carbon dioxide (CO_2), and inorganic ions, through dehydrogenation or hydroxylation [84,85]. There are three main types of AOP, such as electrochemical oxidation, Fenton process, and photocatalytic treatment which are commonly used in oily wastewater treatment [59]. The advanced oxidation methods are associated with high mineralization efficiency, rapid oxidation reaction rates, and minimal toxicity, which are favorable in the treatment of oily wastewater [86,87]. For instance, Gotsi et al. [88] investigated the treatment efficiency of the wastewater generated from olive oil mills using electrochemical oxidation. In this study, a flow-through electrolytic cell with internal recycling at a voltage of 5, 7, and 9 V, NaCl concentrations of 1%, 2%, and 4%, recirculation rates of 0.4 and 0.62 L/s, and initial COD concentrations of 1475, 3060, 5180, and 6545 mg/L were examined. Results showed that increasing voltage, salinity, and recirculation rates led to a high oil removal efficacy. In photocatalytic research, previous studies mostly used TiO_2 and ultraviolet and/or solar light for oil degradation [89,90]. These studies showed good results when oxidizing PAHs, BTEX, and phenols in oily wastewater [81]. In this line, Sivagami et al. [91] examined the treatment of total petroleum hydrocarbons in oil spill sludge using the combination of ultrasound and the Fenton process. Different operating parameters such as pH, ultrasonic power, the weight ratio of hydrogen peroxide to iron [H_2O_2/Fe^{2+}], Fenton reagent dosage, addition of salts, and contact time were investigated. The high petroleum removal efficiency (84.25%) was obtained at a pH of 3.0, sludge/water ratio of 1:100, ultrasonic power of 100 W with 40–50% ultrasonic amplitude, an H_2O_2/Fe^{2+} weight ratio of 10:1, and an ultrasonic treatment time of 10 min. While Liu et al. [81] investigated the treatment of offshore produced water using a photocatalytic ozonation system with TiO_2 nanotube arrays (TNA) and UV-light-emitted diode (UV-LED) irradiation. Results showed that ozone significantly improved the oxidation rates and removed the PAHs within 30 min. However, the high cost of the treatment process, high energy consumption, corrosion problems in treatment facilities, harmful and toxic catalysts, solid waste generation, and complex chemistry are the downsides of AOPs [87,92].

Table 1. Physical methods to treat oily wastewater.

Method	Pros	Cons	Types of Removed Oil	Reference
Gravity Separation	Low cost, simple device	Large footprint, limited separation capacity and poor treatment effect on emulsified oil	Free oil	[11]
Dissolved Air Flotation (DAF)	High-quality effluent, improved surface loading	High operating cost, large footprint	Dispersed oil, emulsified oil	[73,74]
Coagulation	Low cost, small equipment, easy to operate, well-established and practical	Poor treatment effect with surfactant, complicated composition, a large amount of coagulant, generation of sludge	Dispersed oil, emulsified oil	[11,75]
Membrane Separation	High-quality permeate, small footprint, low energy input, low generation of waste	Membrane fouling requires cleaning and backwashing, and incurred cost	Dispersed oil, emulsified oil	[8,11,76]

The adsorption method refers to the physical adhesion of the polluting chemicals onto the surface of a solid, which is used to eliminate soluble oil and chemically stable emulsions [93]. In this method, a wide range of materials such as activated carbon, bentonite, sand, coal, polypropylene, and organoclay are used. In this line, Okiel et al. [94] investigated the treatment of oily wastewater using bentonite, powdered activated carbon, and deposited carbon. The impacts of contact time, adsorbent weight, and the concentration of adsorbate on the oil removal were examined. The oil removal efficiency was improved by increasing the contact time and the adsorbent weight. Deposited carbon and bentonite had higher adsorptive capacities compared to the powdered activated carbon. Deposited carbon and bentonite have a higher porosity and surface area; therefore, they are more suitable for oil removal. In another study, Islam [95] investigated the efficacy of organoclay to remove the oil by measuring its adsorption capacity. It was found that organoclay was effective in removing different concentrations of oil in the oily wastewater. The contact time of 3 h between organoclay and the oil-in-water emulsion was optimal for high oil removal efficiency. The oil removal efficiencies for initial oil concentrations of 750 ppm, 1000 ppm, and 1500 ppm were 28.20%, 35.75%, and 40.036%, respectively. Although adsorption is an effective treatment, the preparation of adsorbents is time-consuming, complicated, and expensive; additionally, as the adsorbent is used there is a reduction in the active ingredient percentage, mechanical strength, and adsorption selectivity, which restricts its application [11,96].

Chemical treatment with demulsifiers (surface active agents) is also used to treat oil-in-water emulsions as the chemical additives accelerating the separation of oil and water [97]. When demulsifiers are used, they move to the water and oil interface, weakening the surface tension and improving coalescence. This method is rapid, efficient, and capable of decreasing oil viscosity in an emulsion and separating oil and water [98,99]. Previous studies showed that the demulsifier's molecular weight had a significant impact on the performance of demulsification. Increasing molecular weight resulted in better separation and there was a linear correlation between the removal efficiency and the molecular weight [100,101]. Razi et al. [102] investigated the impact of various demulsifier formulations on the efficacy of chemical demulsification of heavy crude oil. Results demonstrated that the different surfactant demulsifiers varied in removal efficiency. The formulated surfactant showed a higher efficiency in the demulsification of a medium crude oil emulsion compared to a heavy crude oil emulsion. The different surfactant efficiencies were associated with asphaltene content which was lower in the medium crude oil. Different studies reported the use of demulsifiers formulated from polymers, such as alkene oxides diester, ethylcellulose (EC), Tween non-ionic polymer, and polyester to improve the efficiency of oil removal [103–105]. As an example, the demulsification efficiency of 97.5% was reported within 45 min of the demulsification process using a polyester-based demulsifier. Demulsification mitigates the requirement for heating and retention time for the separation process [98]. The overdose of demulsifiers negatively impacts treatment efficiency by forming a rigid layer caused by aggregating particles; therefore, adding the ideal amount of demulsifier during oily wastewater treatment is required to enable propagation expansion at the interface when it is dissolved in the oil phase [82,99]. Table 2 shows the advantages and disadvantages of chemical methods and the types of oil that can be removed.

Table 2. Chemical methods to treat oily wastewater.

Method	Pros	Cons	Types of Removed Oil	Reference
Adsorption	Depending on the type of adsorbents it has high selectivity, high adsorption capacity, high reuse rate	Material preparation is time-consuming and complex, adsorbing water by organic adsorbents as much as oil adsorption	Emulsified oil	[11]
Electrochemical Oxidation	Low space requirements, efficient treatment in a short time, effective removal of oil and grease	High cost, high power consumption, complex device	Emulsified oil, hazardous metal ions	[11]
Photocatalytic Process	Able to oxidize persistent combinations which are not oxidized during biological treatment	High energy consumption, low efficiency	Emulsified oil	[59,81]
Fenton Process	Effective in removing toxic wastewater, short reaction time, using easy-to-handle reagents	High cost of consuming reagents, harsh acidic atmosphere, high generation of ferric sludge	Emulsified oil	[106,107]
Demulsification	Effective in accelerating the separation of oil and water process, easily used with reasonable cost, minimizing the amount of heat and settling time required	Expensive, toxic, high consumption	Emulsified oil	[108]

2.2.3. Biological Methods

In the biological treatment method, different types of microorganisms are cultured and used to degrade the organic contaminants in wastewater [69]. The microbes use organic matter such as oil as a source of carbon along with sources of phosphorus and nitrogen as nutrients to perform metabolic processes. As a byproduct of microbial metabolism, the oil contaminants are converted to less harmful products such as carbon dioxide, methane, oxygen, and nitrogen gas [109]. Biological studies used activated sludge to treat oily wastewater [9,110–112]; for instance, Shokrollahzadeh et al. [111] achieved 89% and 80% COD and total hydrocarbon removal, respectively, when treating petroleum wastewater. In another study, Sanghamitra et al. [9] reported the removal efficiency of oil ranging from 54.6–80% using an aerobic batch reactor for the treatment of oily wastewater.

Using the biofilm method, layers of microorganisms are grown on a filter material. When the microbes contact raw water, they begin to biodegrade the organic contaminants in the wastewater [11]. The advantages of this method include eco-friendliness, compatibility with carbonaceous stabilization, and it has a cheap and straightforward operation [9]. Sun et al. [113] studied the biofilm-MBR technology used to treat shipboard wastewater. Two processes such as dead-end sidestream and recycle sidestream configurations of a biofilm-MBR were used. A membrane permeate quality of less than 5 mg/L oil was obtained in each process configuration. In this research, a remarkably enhanced membrane performance and better quality of permeate were obtained by recycling the concentrate solution back to the biofilm reactor because of better bio-flocculation and biodegradation of oil compounds in the process. In another study, the impact of biofilm formation on membrane performance was assessed in an MBR unit treating petrochemical wastewater. The biofilm formation in the MBR system enhanced COD removal efficiency by up to 95% [114]. Biological methods have difficulty handling various microbial behaviors under different environmental conditions [69]. The advantages and disadvantages of biological methods are presented in Table 3.

Table 3. Biological methods to treat oily wastewater.

Method	Pros	Cons	Types of Removed Oil	Reference
Microbial Metabolism	Low cost, no additional chemical operation, high removal of BOD and SS	Time-consuming, low efficiency, difficult to handle on a large scale, microbial mechanism complexity	Emulsified oil	[11,76]
Biofilm	Low cost, simple operation, high separation efficiency	Formation of diffusion resistance to the substrate and nutrient as a result of increasing cell layer, immobilization process takes several times at the beginning of the experiment, limited operation time	Emulsified oil	[11,76]

Reviewing the benefits and drawbacks of different physical, chemical, and biological processes demonstrated the characteristics of each technology that need to be considered when selecting a suitable approach to treating oily wastewater. Membrane technology, either in the form of physical separation or MBR, is a suitable solution to treat oily wastewater compared to other methods since it has a selectivity feature, small footprint, high volumetric loading rate, and high effluent quality [3,6,16]. It is important to consider different aspects of membrane technology such as material, fabrication method, and configuration to improve oily wastewater treatment.

3. Membrane-Based Technology

3.1. Membrane Fabrication

When a membrane module is in a direct contact with oily wastewater, the membrane materials should sustain integrity for a long period of time and maintain a good performance based on its chemical resistance, mechanical strength, durability in a wide range of pH levels, heat resistance, hydrophilicity, surface roughness, and its membrane flux [11,115]. Therefore, an important aspect in the preparation of membranes with high efficacy is the specification of a suitable fabrication method and membrane material [116].

The preparation of the membrane is dependent on the required morphology [116]. Some universal methods for membrane preparation to improve performance are phase inversion, interfacial polymerization, stretching, and electrospinning. Polymeric membranes designated for oily wastewater treatment are often prepared using phase inversion or electrospinning [116]. The phase inversion method is a simple and straightforward technique for preparing a membrane [117]. In this process, the thermodynamic state of a homogeneous polymer solution is altered by contacting it with another phase (liquid or vapor), this will improve the formation of a solid phase. This is a common method for the production of microporous polymeric membranes [118]. In contrast to the phase inversion, electrospinning leads to membranes with a relatively uniform pore size distribution with high interconnectivity of pores and significantly higher porosity [119]. Electrospinning continually generates ultrathin polymer fibers of nano to micrometer sizes. Electrospun membranes include a nonwoven and persistent web of nanofibers with an intricate pore structure [120]. The electrospinning method has a simple and versatile instrument and is a continuous, scalable, and cost-effective process [121]. However, the use of organic solvents in the electrospinning method is harmful to the environment and human health [121,122].

Commonly used polymers for membrane material are polyvinylidene difluoride (PVDF), polyethylene (PE), polyacrylonitrile (PAN), and polytetrafluoroethylene (PTFE) [116,123]. Polymeric membranes are hydrophobic, hence they are prone to high rates of membrane fouling, therefore, prior to use, the membranes are typically modified [116]. Polymeric membranes can be enhanced by membrane surface modifications with hydrophilic polymers or by coupling diverse manufacturing techniques to improve the membrane performance. The resulting hydrophilicity not only helps prevent the oil droplets from blocking the membrane surface and improving the treatment efficiency but also saves a significant amount of cost in membrane maintenance and replacement [26,117,124].

The main surface modification methods are surface coating and surface grafting of the membrane [124]. Surface coating is an economical practice for membrane functionalization and can be easily implemented in industrial-scale operations. The coating acts as a protective layer against the harsh oily wastewater environment to prolong membrane life [123–127]. There are different techniques involved such as sulfonation or cross-linking that are used to secure the coating on the membrane surface. Surface grafting forms covalent bonds on the surface of the membrane with new functional groups [123,124]. This type of surface modification also has the potential to expand or shrink membrane pore size. Surface grafting is achieved through chemical processors with high-energy radiation or UV irradiation. The hydrophilicity of the surface is accomplished by grafting polar functional groups on the membrane surface [123,124,128,129]. When the membrane surface is modified, the charge on the surface of the membrane is designed to be the same charge as the foulants in the wastewater that it is treating, resulting in repulsive electrostatic forces to reduce fouling [130,131]. Inorganic nanoparticles such as SiO_2, Al_2O_3, clay, ZrO_2, TiO_2, and ZnO are often used to enhance polymeric composite membranes, and increase thermal stability, permeation, and antifouling properties [23].

3.2. Membrane Selectivity

The separation efficiency of the membranes depends on their selectivity since membranes are semi-permeable barriers through which selectivity between species can be obtained to allow for the separation between unwanted and wanted particles. They will allow the passage of desired species and block the passage of undesirable ones [115]. To achieve selectivity, membrane pore size must be carefully chosen [115,132]. The type of wastewater to be treated dictates the best membrane pore size to be used [26]. Based on the molecular weight cut-off (MWCO), membrane filtration is divided into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) [133]. The largest pore size ranging from 100–1000 nm belongs to MF which is capable of separating suspended solids and bacteria through the mechanism of convective pore flow [134]. The pore size of UF is smaller than that of MF ranging from 5–100 nm. UF can be used in the filtration of color, viruses, aroma, and colloidal organic substances [135]. NF has a pore size of 1–5 nm, leading to higher organics removal than UF membranes [134,136]. RO is characterized by a pore size of 0.1–1 nm and is capable of removing salinity from wastewater [137]. MF and UF membranes have been extensively reported for oily wastewater treatment, and, among these membranes, UF is the most favorable due to a low-pressure operation, and low capital and operating costs [10].

3.3. Membrane Configuration

Configuration is another factor that influences the application and effectiveness of the membrane when treating oily wastewater. Membrane configuration refers to the geometry of the membrane and its position in space in relation to the flow of the feed fluid and the permeate. As most industrial membrane installations are of modular design, membrane configuration also determines the manner in which the membrane is packed inside the modules [138]. The desired characteristics of a membrane configuration include compactness (i.e., the capability of packing as much membrane surface as possible into a module), low resistance to tangential flow (i.e., low friction and energy consumption, low-pressure drop along the retentate flow channel), uniform velocity distribution, easy cleaning and maintenance, and low cost per unit membrane area [138].

Two process configurations in membrane technology are submerged and sidestream which are used in the treatment of oily wastewater [139,140]. In the submerged type, the membrane is located inside the membrane tank while in the sidestream, the membrane is placed outside the membrane tank [141]. Submerged configuration is easy to operate and needs low energy compared to the sidestream setup, however, the cleaning process in the submerged is more complicated than that of the sidestream [142]. Three strategies can be used in submerged processes to limit fouling, such as increasing the aeration flow rate, decreasing the membrane flux, and using physical or chemical cleaning. Aeration produces a shear force on the membrane surface through the rise of coarse bubbles which mitigates the accumulation of foulants on the membrane surface. Using lower membrane flux limits membrane fouling since it decreases the rate of foulants reaching the membrane surface. Implementing the mentioned strategies mitigates the need for membrane cleaning. Using physical cleaning is more straightforward than chemical cleaning since chemical cleaning requires chemicals and generates chemical waste. Often, physical cleaning is enough to remove the foulants on the surface of the membrane (reversible fouling). Chemical cleaning is seldom needed and is mainly used to unclog the pores of the membrane (irreversible fouling) [143,144].

In the sidestream configuration, the mixed liquor from the bioreactor is circulated on the membrane surface at high crossflow velocities and pressures, requiring high energy demand to mitigate membrane fouling [145]. This configuration, however, experiences more severe membrane fouling than submerged membranes due to the lack of sufficient shear force on the membrane surface. Aeration is much more effective at producing a high shear force on the membrane surface than circulating mixed liquors at high crossflow velocity [144,146].

There are four main membrane configuration categories, such as plate and frame, tubular, spiral wound, and HF. The plate and frame is one of the earliest modules produced [142] which contains two flat-sheet membranes that are stretched across a thin frame. There is a vacuum space between the two membranes, which supplies the driving force for filtration. Many plates are organized in a cassette to expand the surface area. The flow for the immersed cassette is designed to be from the outside-in (i.e., the air stimulates liquid to be crossflow along the plates). This configuration has a limited application and is mostly used for treating wastewater with high amounts of SS [142]. The process generates turbulence and hampers cake formation, and, therefore, reduces membrane fouling. The crossflow of air helps dissolve oxygen in the membrane tanks and mixes wastewater in the reactor [147–149]. The packing density of plate and frame ranges between 148–492 m^2/m^3 which is lower than spiral wound and HF. This membrane has a moderate potential for membrane fouling, and its cleaning is easier than spiral wound and HF, however, its manufacturing cost is high.

The tubular membrane contains outer housing (i.e., shell) which is tubular. A perforated or porous stainless steel or fiberglass pipe is placed within the tubular shell, and a semi-permeable membrane is inserted within the stainless steel or fiberglass pipe. The permeate passes through the pipe into the inside of the housing and is collected by the permeate outlet [138,142,150]. The packing density of the tubular membrane is between 20–374 m^2/m^3 which is the lowest compared to other configurations. This type of membrane maintains a high tangential velocity in the feed and is used for feed containing a high amount of SS [138,142]. This membrane has a low potential for membrane fouling and its cleaning is the easiest due to the large diameter, however, its manufacturing cost is high [138].

Spiral-wound membranes are comprised of the membrane, feed spacers, permeate spacers, and a permeate tube. Wastewater moves tangentially across the flow channel along the length of the spiral-wound membrane. The water will cross the membrane surface, rejecting larger particles, through the permeate spacer, and into the permeate tube. The permeate will exit at the other end of the spiral-wound membrane and into a separate tank [151,152]. The packing density of the spiral wound is 492–1247 m^2/m^3 which is lower than HF. This membrane has a high potential for membrane fouling, is hard to clean, and has moderate manufacturing costs [142].

HF membranes are made up of long threads or fibers of hollow membranes. The tubes are thin with diameters ranging from 1 mm down to capillary size. The membranes are installed on a supporting structure that functions as a manifold for permeate transport and a system for air distribution. Similar to the plate and frame module, HF relies on aeration to avoid excessive cake layer formation on the membranes. This type of membrane generally functions in an outside-in manner, where the fibers have a vacuum and the water flows from the reactor to the inside of the hollow fibers and out of the system [138,147]. This configuration is capable of housing large membrane areas in a single module. The packing density of HF is 492–4924 m^2/m^3 which is the highest and the manufacturing cost is low [142]. Table 4 shows the comparison of different membrane configurations.

Table 4. Comparison of different membrane configurations.

Configuration	Applications	Advantages	Disadvantages	Oil Removal Efficiency (%)	Reference
Plate and Frame Membrane Module	UF and RO, MBR, Food and beverage, Oily Wastewater	Easily removing solids from water, easy to clean, moderate potential for fouling	Low packing, high cost, not back flushable, the lowest membrane area per unit volume, low efficiency compared to other configurations, high-pressure drop	Hybrid MF/UF: 99.9% UF: >95%	[138,140,142,147,153,154]
Tubular Membrane Module	MF/UF, wastewater with high dissolved and suspended solids, oil, and grease	Less fouling compared to plate and frame, handling the highest solids load, easy to clean	Low packing density, not back flushable, very high cost, very large footprint	UF: 99% UF: 98.04%	[142,147,153,155,156]

Table 4. *Cont.*

Configuration	Applications	Advantages	Disadvantages	Oil Removal Efficiency (%)	Reference
Spiral-Wound Membrane Module	RO/NF/MF UF, whey protein concentration, lactose concentration, cathodic/anodic paint recovery, dye desalting, sulfate removal, oil separation	Easy cleaning through cleaning in place, small footprint, robust design, low capital and operating cost	Lower packing density than HF, high potential for fouling, not back flushable	UF: 90.1% UF: 99.7%	[35,142,153,157–159]
HF Membrane Module	MF/UF and RO, MBR, industrial wastewater, oily wastewater juice processing, biotech applications	Moderate capital cost, very high packing density, back flushable, capable to generate movement by mechanisms such as bubbling, higher membrane area per unit volume compared to flat-sheet membranes	Fiber breakage, high operating cost, high potential of fouling	UF: 99% UF: 98.5%	[142,147,153,158,160,161]

4. HF Membrane Module and Its Performance Parameters

HF membrane application has caught a great deal of attention because they are able to separate and treat a wide range of complex wastewater, such as oily wastewater [139,160,162–164]. HF membranes are desired over other membrane configurations because they are compact modules with very high membrane surface area, high packing density, the feasibility of backwashing, self-supporting structure, simplicity of handling, and low manufacturing cost [23,24,142,149,165]. Three typical fiber packing configurations used for full-scale submerged HF membrane modules are the curtain fiber bundle, cylindrical fiber bundle, and cylindrical bundle with free fiber end, as shown in Figure 1. The most common module packing configuration is the curtain-type fiber bundle. This sort of packing configuration generally consists of 4 to 12 fiber sheets and permits well-defined fiber spacing and high tank packing density [166]. The cylindrical fiber packing configuration can attain high module packing density; however, the general tank packing density is typically less than the curtain type modules. The cylindrical with the free fiber end configuration can only be permeated from one end of the module, which could consequently increase the non-uniform distribution of TMP along the fibers, specifically when small-diameter fibers are used. Modules with the same packing configuration are compiled into a cassette to create an engineering module unit with a common permeate collection conduit to increase packing density, and, consequently, the productivity of a membrane tank [166].

Since membrane fouling is a major issue in membrane filtration technology, it is required to investigate the main parameters affecting fouling in HF membranes. Membrane properties (i.e., fiber diameter, length, and tightness), hydrodynamic conditions (i.e., surface shear, air flow rate), feed properties (i.e., foulant properties, concentration, viscosity), and operating conditions (i.e., temperature, membrane flux) affect the membrane performance [23]. Previous studies showed that low membrane fouling is a result of a low HF packing density, through having few fibers, widening the HF module, and changing the module configuration [167,168]. In addition, designing modules with improving lateral flow or lateral movement of the fibers can enhance the submerged HF systems [169]. Yeo and Fane [167] concluded that a single fiber operated better than a multi-fiber HF module in the absence of aeration because of module blocking. However, fiber movement as a result of aeration is another factor that affects the fouling deposition on HF membranes. Bérubé and Lei [170] reported that a multi-fiber module outperformed a single fiber in the presence of aeration due to inter-fiber interactions which cause mechanical erosion of the foulant layer. Pourbozorg et al. [171] studied the effect of vibration induced by perforated plates on the fouling reduction of submerged HF membranes. Results showed that the fouling rate was dependent on kinetic energy and eddy length scale; greater turbulence kinetic energy resulted in a lower fouling rate.

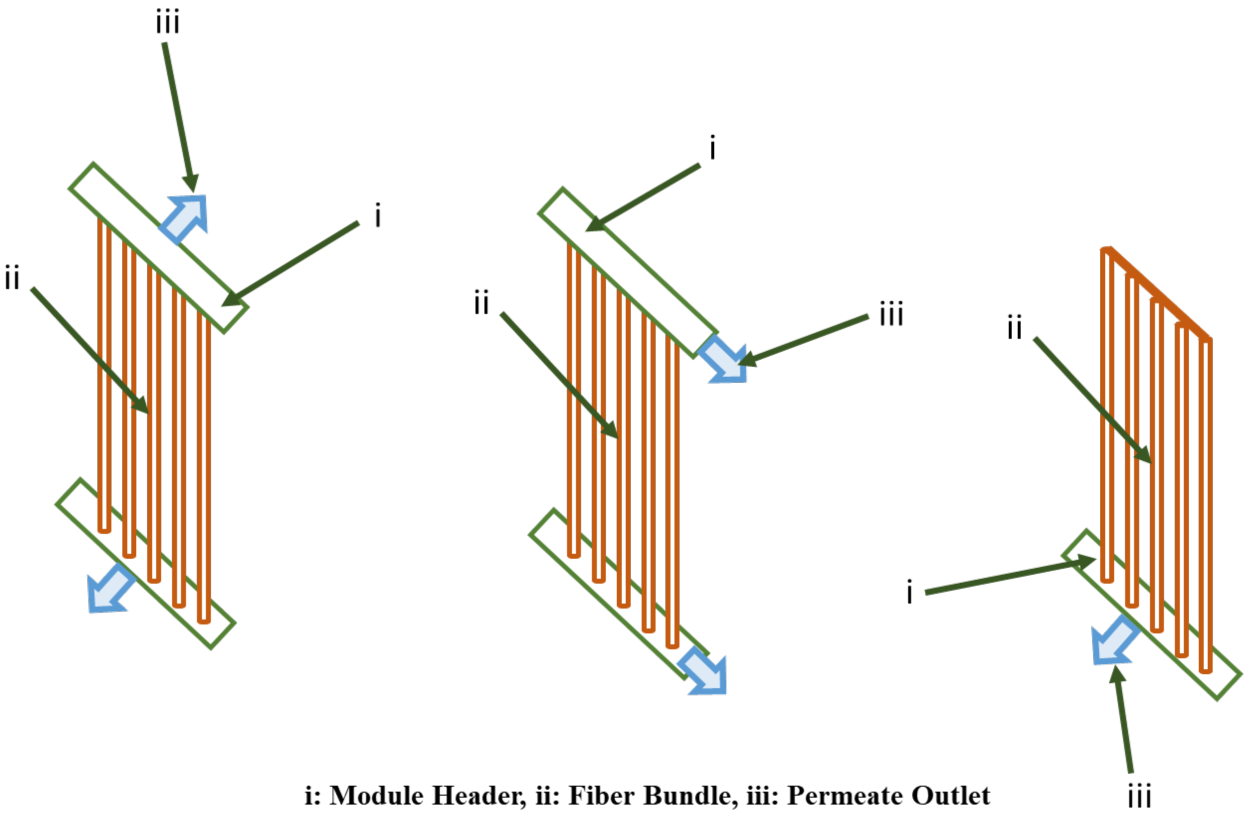

Figure 1. Various fiber packing configurations of submerged HF membrane module "adapted from [166]".

Higher fiber movement obtained by using looser fibers improves the back-transport of the foulants from the membrane caused by physical contact among the fibers, decreasing membrane fouling [172,173]. A study reported that tight fibers in HF led TMP to increase 40% faster compared to fibers with a 4% looseness [174].

Previous experimental studies showed that a smaller HF diameter enhanced the performance of submerged systems as a result of higher fiber mobility under aeration [172,174,175]. In addition, long fibers (i.e., length in the range of 0.3–1 m) reduced membrane fouling due to non-uniform deposition of particles and high movement due to aeration [176,177]. Recently, Khanafer and Assad [178] reported that the increase in fiber length from 1 to 1.5 m led to an increase in permeate productivity by approximately 248%. They concluded that fiber length and inner radius should be optimized to enhance productivity and filtration uniformity in HF systems.

In terms of hydrodynamic conditions, crossflow velocity, aeration, and vibration in submerged HF systems impact surface and unsteady-state shear which affect membrane fouling and performance [179–182] Previous studies concluded that aeration intensity enhances the hydrodynamics of the membrane filtration system; air bubbles change the structure of the fouling layer, and decrease specific resistance [181,183]. Selecting an optimum airflow rate was crucial for treatment efficiency; a higher aeration flow rate than the critical value has shown to have no effect on system performance [184,185]. The position of aerators in the HF membrane filtration system was also studied and results showed that the injection of air at the bottom of the membrane fiber enhanced the overall system performance [186].

In MBR technology, parameters such as food-microorganism (F/M) ratio, SRT, HRT, and aeration flow rate have a significant impact on HF membrane performance. The operational parameters have a substantial effect on microbial extracellular polymeric substance (EPS) and soluble microbial products (SMP) production. EPS is defined as an extracellular polymeric substance of biological origin that participates in the formation of microbial aggregates [187]. SMP is a soluble microbial product that is comprised of hydrolysis products of EPS and decay products of active cells [188].

For microbes to effectively biodegrade hydrocarbons a balance between food entering the bioreactor and the microbes in the bioreactor is necessary. With a high F/M ratio, the bacteria are scattered within the bioreactor and reproduce quickly as a result of having more food than microorganisms in the enclosed environment. Due to the dispersion of bacteria, large flocs are unable to form, leading to poor settling [189]. Additionally, at high F/M ratios, an increase in membrane fouling occurs due to high food utilization by biomass resulting in increased EPS and SMP production [190–192]. As an example, Dvořák et al. [193] investigated the impact of the F/M ratio on membrane fouling and EPS production. Results showed that a high F/M ratio led to a high concentration of EPS. The high F/M ratio disturbs the balance between the food supply and the mass of microorganisms in the system, leading to higher metabolic activity and microbial growth [191,192].

At low F/M ratios, the food supply is restricted, resulting in the bacteria producing a thicker slime layer, losing their motility, clustering together, and forming large flocs that settle easily. At high sludge age and mixed liquor suspended solid (MLSS) concentration, the system demands fewer nutrients, owing to the decrease in excess sludge production. The MBR system reaches an equilibrium where the nutrient provided matches the microbial maintenance demand [189,194]. Therefore, it is favorable to use a low F/M ratio to mitigate membrane fouling.

SRT is the amount of time that the sludge is retained in the membrane bioreactor [139]. The key to effective wastewater treatment is to maintain a high MLSS concentration as a result of high sludge age. A high sludge age can be attained through long SRT, which allows the retention of particulate, colloidal, and higher-weight organics, giving the microbes maximum chance to degrade organic compounds, and allows for the acclimation of microbes to the biodegradable compounds [189]. However, extremely high SRTs are not desirable as they increase membrane fouling due to the accumulation of biomass (high MLSS) and increasing sludge viscosity [190], therefore, it is important to periodically discharge some of the activated sludge to maintain desired conditions within the membrane bioreactor [195]. Low sludge age and high SRT exert high stress on the bacterial community, resulting in high EPS and SMP production [180].

HRT is the amount of time that the wastewater remains in the bioreactor for biological degradation [57]. Long HRTs allow for a longer contact time of the microbes with the organic compounds in the wastewater; thus, increasing the removal efficiency of the contaminant and COD in the wastewater. However, long HRT is only effective provided that the MBR system is conducted under steady-state conditions (i.e., the microbial community is acclimated to the wastewater and high sludge age is achieved) [189]. Short HRT increases the organic loading rate, which puts a stain on the bacteria. The bacteria then produce excess EPS and SMP which contribute to membrane fouling and membrane flux decline [189,190]. The optimal HRT is dependent on the types of bacteria in the activated sludge and wastewater characteristics [190]. For example, Yang et al. [169] tested HRTs of 1, 2, 4, and 8 h to treat real domestic wastewater in a membrane filtration system. The HRT of 8 h had the best COD removal efficiency and had the lowest rate of TMP increase resulting in the lowest COD removal efficiency as well as the highest rate of TMP increase. Razavi and Miri [139] studied the treatment of HF-MBR to treat real petroleum refinery wastewater and tested HRTs of 25, 30, and 36 h. They concluded that the lowest removal efficiency and highest membrane fouling were obtained at HRT of 25 h and the best membrane performance occurred at HRT of 36 h.

Aeration provides oxygen to microorganisms, offers a homogenous distribution of activated sludge and fluid, and decreases fouling on the surface of the membrane [196]. Deng et al. [197] showed that increasing aeration changed the total quantity and composition of SMP [198]. Meng et al. [199] investigated the impact of different aeration flow rates such as 150, 400, and 800 L/h on membrane fouling in three MBR systems. The results showed that at a high aeration flow rate (800 L/h) colloids and solutes were the dominant foulants because of the breakup of sludge flocs. Using a moderate aeration flow rate was suggested to keep the sludge flocs intact so as not to disturb the sludge filterability, thus, resulting in a decrease in membrane fouling and an enhancement of membrane flux [200].

5. The Application of HF Membrane in Oily Wastewater Treatment

HF membrane has been used in two ways including physical separation and the integration of physical separation and biological method (MBR). The first section focuses on research works that solely use HF membrane filtration, and the second section reviews MBR for the treatment of oily wastewater. The selected studies will help us to understand the strengths and weaknesses of HF membrane technology, and the knowledge gaps in this field. The chosen studies were among the most highly-cited published HF research works, focusing on oily wastewater treatment.

5.1. Physical Separation Studies

Membrane surface modification is an effective method for improving membrane performance. Membranes are modified to be more hydrophilic; a higher affinity of water to hydrophilic membrane surface can lead to the development of a hydration layer on the membrane surface, which decreases membrane fouling by preventing hydrophobic components, such as oil, from attaching to membrane surface [163]. For example, Zhu et al. [163] constructed a new HF membrane using dry-wet spin phase inversion and modified it with hydrophilic and oleophobic surface features through blending a polymer, P(VDF-co-CTFE)-g-PMAA-g-fPEG, with a PVDF membrane to treat oily wastewater containing hexadecane, crude oil, and palm oil. The oil contact angle of the unmodified membrane was approximately 15° or lower which showed the high oleophilicity of the membrane while modified membranes had an oil contact angle of about 75° indicating a higher oleophobicity. Therefore, an unmodified membrane adsorbed a higher concentration of oil compared to a modified membrane due to its hydrophobicity. Results showed that the flux decline of unmodified membranes reached about 88% but modified membranes were at around 17%. The modified HF membrane had a greater water flux (i.e., 72 $L/m^2 \cdot h$) during oily wastewater treatment. The flux recovery rate of the unmodified membrane after cleaning with deionized water was 40%, while the flux recovery rate of modified membranes was more than 89%, which was attributed to the oleophobic property. In addition, more than 98%, 98%, and 70% oil removal efficiencies for oily wastewater containing hexadecane, crude oil, and palm oil were achieved, respectively.

Luo et al. [201] used a novel sulfonated polyphenylenesulfone (PPSU) polymer with a super-hydrophilic feature to fabricate triangle-shaped tri-bore HF-UF membranes using a dry-jet wet-spinning process. Three membranes, polyphenylenesulfone, sulfonated polyphenylenesulfone with sulfonation degree of 1.5 mol%, and sulfonated polyphenylenesulfone with sulfonation degree of 2.5 mol% were compared. The oil contact angle for the unmodified membrane was 89.6° while for modified membranes with a sulfonation degree of 1.5 mol% and 2.5 mol% were 111.9° and 115.69°, respectively. This indicates that the modified membranes were more hydrophilic than the unmodified ones. Results showed that the unmodified membrane had a permeate flux decline of 82.1% and the total resistance increased by 5.7 times, while in modified membranes, the permeate flux decline was less than 55% and the total resistance increased only by 1.7–2.2 folds. The membrane with 1.5 mol % showed the highest flux and all membranes had 95.4% of TOC removal efficiency.

Otitoju et al. [202] compared three different membranes under the same operating conditions. The three membranes were PES, PES/SiO_2, and tetraethyloxysilane PES/(TEOS). All the membranes were prepared using dry-wet spinning. The oil contact angle of the PES/TEOS membrane was 125.47° which was the highest compared to PES and PES/SiO_2. PES/TEOS showed the best hydrophilicity compared to the other two membranes due to the formation of a large-scale network of Si-OH or hydroxyl groups originating from TEOS on the membrane surface. The PES/TEOS membrane had a 99.98% oil removal efficiency and a permeate flux of 90.937 L/m^2·h, while PES/SiO_2 had an oil removal efficiency of 97.48% and permeate flux of 74.856 L/m^2·h, and PES had an oil removal efficiency of 95.77% and permeate flux of 60.112 L/m^2·h. Therefore, the unmodified membrane (PES) had the lowest oil removal efficiency and permeate flux compared to the modified membranes. This study also found that the modified PES/TEOS membrane showed exceptional antifouling properties and the oil deposition on it was easily washed through physical cleaning.

The blending of the main polymer with a hydrophilic polymer in the casting solution is a significant alternative, decreasing the fabrication cost compared to the other surface modification methods. In this line, Johari et al. [164] treated oily wastewater using fabricated HF membranes (i.e., a combination of hydrophilic polyamide imide (PAI) with three ratios of sulfonated poly (ether ether keton) (SPEEK)) through a phase inversion process. The oil contact angles in the unmodified membrane and modified membranes such as PAI/SPEEK 95/5 and PAI/SPEEK 85/15 were 105.7°, 111.7°, and 121.82°, respectively. This showed that the modified membranes were more hydrophilic than the unmodified ones. Results showed that oil removal efficiency was over 95% and the modified membrane showed a permeate flux of 39.6 L/m^2·h which was 2.5 times the original membrane flux. Shen et al. [203] prepared a hydrophilic HF membrane with an antifouling feature by grafting sulfobetaine methacrylate (SBMA) onto polysulfone (PSU). The tests were conducted for six different types of oils, soybean oil, olive oil, lard oil, gasoline, diesel oil, and crude oil. The oil contact angle of the modified membrane was more than that of the PSU membrane which showed a better antifouling feature of the modified membrane. The modified membrane had a higher flux for oil/water emulsion of two vegetable oils (130–220 L/m^2·h) than the original membrane. When oil concentration was at 1000 mg/L, the oil removal efficiency was over 91.1%. The flux was completely recovered after cleaning the membrane. This new method of membrane fabrication can operate efficiently to remove different oil compounds in wastewater.

El-badawy et al. [204] treated oily wastewater containing crude oil using PVDF-PET braid-reinforced HF membranes created with very thin and uniformly coated hydrophilic/oleophobic material through a dry-jet wet spinning process. Results showed that the flux reached 620 L/m^2·h and oil removal efficiency was 88% due to high porosity and underwater oleophobicity. The modified membrane showed excellent antifouling features against wastewater containing crude oil with a high flux recovery of more than 95% for three filtration cycles.

Since producing a hydrophilic membrane is costly, membranes that are fabricated from low-cost and green materials and have promising results in terms of oil/water separation are desired. Baggio et al. [205] developed amphiphilic membranes from polyethylene terephthalate (r-PET) and chitosan. This is a hydrophilic and biodegradable polymer manufactured by deacetylation of chitin. The membrane was fabricated through an electrospinning method using environmentally-friendly materials. The flux ranged between 512–991 L/m^2·h. Results showed that the membrane removed over 95% of heavy and light crude oils from wastewater. In this study, the use of biodegradable polymers such as chitosan is beneficial due to their non-toxic feature, availability, and inexpensiveness.

Previous HF physical separation studies demonstrated that the modification of membranes using appropriate materials has a drastic effect on oily wastewater treatment, significantly improving permeate flux, oil rejection, and facilitating membrane cleaning. The summary of HF physical separation studies for the treatment of oily wastewater is shown in Table 5.

Table 5. Summary of physical separation studies using modified HF membranes for oily wastewater treatment.

Membrane Material	Additive Polymer	Membrane Pore Size (µm)	Membrane Surface Area (m^2)	Flux ($L/m^2 \cdot h$)	Removal Efficiency (%)	Wastewater Type	Reference
PVDF	PET	0.075–0.401	-	620	above 99.5%.	Crude Oil	[204]
PET	Chitosan	-	-	512–991	>95	Kerosene, hexane, carbon tetrachloride (CTC), and tetrachlo roethylene (TCE)	[205]
Polysulfone (PSU)	Sulfobetaine methacrylate (SBMA)	-	-	267	>98.5	Soybean oil, olive oil, lard oil, gasoline, diesel oil, and crude oil	[203]
Polyethylene terephthalate (rPET)	Polydimethylsiloxane (PDMS)	-	-	20,000	>98%	Oil	[206]
Polyamide imide (PAI)	Sulfonated poly (ether ether keton) (SPEEK)	0.012, 0.03, 0.081	-	32	>95	Petroleum Refinery	[164]
PES	Tetraethyloxysilane (TEOS), polyethylene glycol, silicon sol, and 1-methyl-2-pyrrolidone (NMP)	0.102	0.008	90.937	99.98	Crude Oil	[202]
Polyphenylenesulfone	Sulfonated polyphenylenesulfone (SPPSU)	0.0109–0.0186	-	-	TOC: > 95.4	Oil-in-water Emulsion	[201]
PVDF	P(VDF-co-CTFE)-g-PMAA-g-fPEG	0.097–0.141	0.0085	10–72	98, 99, 70	Hexadecane, Crude Oil, Palm Oil	[163]

5.2. Integration of Physical Separation and Biological Method

In MBRs, the bioreactor acts as a biological treatment processor and the membrane is used as a filter in the filtration process [26]. MBR studies for the treatment of oily wastewater are summarized in Table 6.

In the oil and gas industry, Razavi and Miri [139] used a bench-scale HF-MBR to treat real petroleum refinery wastewater under various HRT, flux, temperature, and different operational conditions. This system was operated at a COD concentration of 580 mg/L and TSS of 110 mg/L. MLSS ranged from 3 to 6.6 g/L with the addition of hydrocarbon in the wastewater and had HRTs ranging between 25–36 h. In the middle of the process, COD and BOD_5 removal increased significantly due to good biomass growth conditions in the system. Reducing HRT during this experiment led to decreasing BOD_5 and COD removals and excess biomass production that led to high membrane fouling and decreased flux. At high HRT, 36 h, the results indicated that the removal efficiency of COD and BOD_5 were 82% and 89%, respectively.

Capodici et al. [207] developed a bench-scale HF-MBR unit to treat synthetic wastewater containing diesel fuel. The membrane flux was maintained at almost 15 $L/m^2 \cdot h$ and HRT was equal to 27 h. A reduction in suspended biomass occurred until day 54 due to exerting stress on the biomass by hydrocarbons and reducing metabolic activity that was not completely acclimated to the substrate. This hindered the production of EPS in the system. Sodium acetate was added to the feed solution which increased the suspended biomass up to 7 g TSS/L and provided an appropriate acclimation level and increased EPS leading to an increase in membrane fouling. However, during the last period, membrane fouling decreased due to unstable oil layer formation on the surface of the membrane which was significantly removed by high crossflow fluid. Removal efficiencies of TPH and total COD were achieved by over 85%.

To evaluate the performance of HF-MBR in saline conditions, Di Bella et al. [208] used a bench-scale HF-MBR system to treat shipboard slops and investigated the effect of salinity and organic loading rates on the system performance in two phases (i.e., phase 1: the acclimation of biomass to salinity was studied, phase 2: the acclimation of biomass to real slops was investigated). Results showed a good biomass acclimation to a gradual salinity (increased up to 15 g NaCl/L) offered the potential development of halotolerant bacteria. The oil removal efficiency was about 50%. In this study, in the first phase, microorganisms were exposed to salinity, and this increased SMP production due to the release of organic cellular constituents through secretion and cell autolysis which led to an increase in membrane fouling. In the second phase, the EPS concentrations decreased slightly, and, therefore, membrane fouling did not increase due to an inhibitory impact caused by the hydrocarbons on the suspended biomass activity. This study showed that a gradual increase of salinity and hydrocarbon is necessary to improve biomass acclimation if bacteria are not halophilic.

In another study, Cosenza et al. [209] reported the treatment of synthetic shipboard slops with two separate bench-scale MBR setups, one with and one without a saline environment. TPH and COD concentrations of the oily wastewater were 20 and 500 mg/L, respectively, at the beginning of the study. In the MBR experiment with a saline environment, the heterotrophic biomass was negatively influenced due to salt shock. During the operation, after day 27, membrane fouling increased which was a result of the stress exerted by the salinity on the biomass that was not acclimated to a substrate which resulted in the release of high SMP concentration. In the MBR experiment without a saline environment, biomass had higher performance efficiency. The ammonium removal efficiency was 70% for both systems, and COD removal efficiencies for MBR experiments with salinity and without salinity were 81% and 87%, respectively. This study shows the importance of acclimating the biomass to high salinity, otherwise, it hinders their ability to degrade contaminants in the wastewater. Another option would be to use halophilic/halotolerant bacteria to obtain high biodegradation performance.

Table 6. Summary of HF-MBR studies for the treatment of oily wastewater.

Scale	Membrane Material	MLSS (g/L)	Pre-Treatment	Membrane Pore Size (μm)	Membrane Surface Area (m^2)	Flow Rate (L/h)	Flux ($L/m^2 \cdot h$)	Air Flow Rate (L/min)	HRT (h)	SRT (d)	Operation Time	Salinity (g/L)	Removal Efficiency (%)	Wastewater Type	Reference
Bench	PVDF, manufactured by Zenon Environmental Systems Inc.	7.6	Electrocoagulation	0.035	0.047	-	12	30	-	-	12 d	-	Oil: 95	Hypersaline oilfield produced water	[210]
Bench	UF, manufactured by ZeeWeed	4	De-oiling, coagulation, flocculation	0.04	0.09	0.8	15	-	27	-	215 d	Conductivity: 1.6 mS/cm	Oil: 85	Synthetic oily wastewater (shipboard slop)	[207]
Bench	UF, manufactured by ZeeWeed	4	Chemical-physical pre-treatment	0.04	0.093	-	15	-	27	-	90 d	0	Oil: 95	Synthetic shipboard slops	[209]
Bench	16 wt% new polyvinylchloride (PVC) and 84 wt% Dimethylacetamide (DMAC) solvent	1	Gravity separation and DAF	0.12	0.00113	-	-	-	-	-	5 d	-	Oil: 100	Oil refinery wastewater	[211]
Bench	Self-made membrane, polypropylene	3–6.6	-	0.15	0.39	0.47	1.205	70	36	-	-	-	COD: 82	Real petroleum refinery	[139]
Bench	Polyetherimide, MF	-	Coalescer bed	0.4	0.5	-	-	-	-	-	8 h	-	Oil: 93–100	Oil produced water	[212]
Bench	PVDF, UF, manufactured by ZeeWeed	-	-	0.04	-	0.5	-	-	-	-	210 d	-	COD: 91.8	Oil refinery wastewater	[213]
Bench	MF, manufactured by Zena Membranes	3.8	-	0.1	0.18	-	10	7	-	Infinite	121 d	8.7 ± 1.7	Oil: 85	Produced water	[160]
Pilot	Coated with LiCl and TiO_2, PVDF, UF	4.5	-	0.034	0.0184	-	82.95	0.0022	4.61	-	-	-	COD: 90.8	Refinery wastewater	[214]
Bench	Polysulfone, manufactured by Polymem-Polymer	9	PAC addition	0.2	0.1	-	2	-	24	20	-	0.09	COD: 96	Effluent from the oil industry	[215]

Table 6. *Cont.*

Scale	Membrane Material	MLSS (g/L)	Pre-Treatment	Membrane Pore Size (µm)	Membrane Surface Area (m^2)	Flow Rate (L/h)	Flux ($L/m^2 \cdot h$)	Air Flow Rate (L/min)	HRT (h)	SRT (d)	Operation Time	Salinity (g/L)	Removal Efficiency (%)	Wastewater Type	Reference
Bench	Polyetherimide, manufactured by PEI, Ultem 1000, GE	-	Sand filter	0.15 ± 0.09	2.78×10^{-2}	2.5	15.82	-	10	-	33 d	-	COD: 67	Oil refinery wastewater	[216]
Bench	PVDF	14–28	-	0.06	0.020	-	6	0.1	10	Infinite	71 d	-	Oil: 98	Industrial oil contaminated wastewater	[217]
Pilot	PVDF, MF, manufactured by Zenon	-	Oil/water separator, floatation system, sand filter	0.04	70	-	-	-	-	-	6 months	0.56	COD: 84	Refinery wastewater	[218]
Pilot	PVC/Alloy manufactured by Litree Co.	-	Aeration tank, air flotation, sand filter	0.006	40	-	-	-	-	-	-	-	Oil: 99	Oilfield wastewater	[219]
Bench	Self-made membrane, polypropylene, sealing procedure with a proper resin, symmetric MF	8.2	-	0.4	0.2	-	0.42	-	31.8	-	11.25 d	-	COD and hydrocarbon: >90	Industrial wastewater containing hydrocarbons	[220]
Pilot	Unmodified	-	Gravity oil separation	-	-	20.82	15	-	-	11	-	-	Ballast water COD: 38, Bilgewater COD: 56	Oily wastewater including ballast and bilge water	[221]
Bench	MF, manufactured by Mitsubishi Rayon Co., Ltd.	9.84	PAC Addition	0.1	0.42	-	3.57	-	4	50	58 d	-	Oil: 99.9	Oily wastewater from gas station	[222]

6. Discussion and Recommendations

The development of HF membrane technology is promising for oily wastewater treatment. Previous studies have indicated that the key challenge in achieving an outstanding membrane performance to treat oily wastewater is to tackle membrane flux reduction and permeability as a result of fouling. Although a lot of studies have been focused on this subject, there may be opportunities for further improvement such as identifying optimal operating parameters to achieve sustainable flux in membrane filtration systems when treating oily wastewater.

Previous research papers discussed the effect of changing HF packing density, fiber length, fiber looseness, and fiber diameter on HF membrane performance. Low HF packing density, long and loose fibers, and small HF diameter allow lateral movement of fibers, which enhances the HF membrane filtration performance.

Inducing shear stress, unsteady-state condition on the membrane surface, and HF fiber oscillation through aeration mitigates membrane fouling and improves the membrane performance. Although aeration is a proven technique for reducing membrane fouling, there is a need for more research on ideal aeration conditions such as optimal bubble diameter, to improve membrane performance, and aeration frequency to reduce the energy demand of the system [13,23]. Bubble diameter affects shear stress on the membrane surface, and, so, finding the optimal bubble diameter will help maintain effective foulant removal and the homogeneity of the mixed liquor, which benefits the biodegradation process in MBR. Until now, there have not been any conclusive studies on the best bubble diameter to improve membrane system performance, therefore, further research on this topic can greatly improve the efficacy of membrane technology.

Previous HF physical separation studies focused on membrane modification to improve membrane performance. Most polymers which have been used in HF membrane production are hydrophobic, and, consequently, they repel water, promoting the passage of oil through the membrane. Oleophilic features allow for membrane pore blockage, the accumulation of an oily cake layer on the membrane surface, and, consequently, a reduction of membrane permeation performance, and decreasing membrane lifespan. Surface modification has proven to be exceptionally effective at preventing oil droplets from breaching the pores of the membrane and maintaining a high flux of water [116,124]. It was observed that membranes modified either by coating or surface grafting had better oil removal efficiencies than membranes that were not modified [3,116]. However, these studies are restricted to bench-scale laboratory experiments, and the challenge remains in preparing modified membranes for commercial use [13,223]. Developing more effective methods to implement membrane surface medication on a large scale would broaden the application of HF membrane technology, particularly in oily wastewater treatment. For example, the expansion of HF membrane application to onsite oil spill treatment or its use as a stand-alone system to treat complicated industrial emulsified oily wastewater. Additionally, recent studies have indicated the potential of using green technology in constructing highly efficient HF membranes [205,206]. During membrane surface modification, sustainable and cost-effective methods through environmentally-friendly materials should be considered [224].

In MBR applications, membrane fouling is still a major challenge similar to physical separation studies which impedes the achievement of excellent membrane performance for the treatment of oily wastewater [225,226]. Factors that affect membrane fouling are operating parameters such as HRT, SRT, and aeration that impact sludge characteristics [116]. A low F/M ratio is required for microbial maintenance, high F/M ratio causes the dispersion of bacteria and results in more EPS and SMP production leading to membrane fouling. Long SRT with high sludge age leads to a better performance of the MBR unit as it allows for biomass acclimation to the wastewater [146]. Longer HRTs improve effluent quality as it gives time for the biomass to degrade unwanted compounds in the water. Aeration flow rate is a vital parameter to monitor the treatment of wastewater since it provides dissolved oxygen to bacteria so that they may perform biodegradation [181]. Low aeration leads to anaerobic conditions in the bioreactor, resulting in the death of aerobic bacteria. A high aeration flow rate contributes to low sludge filterability by breaking floc formations. Both conditions will result in high EPS and SMP in the system which contributes to the clogging or blocking of the membrane.

High salt concentration and the existence of hydrocarbons in feed water cause environmental stress on the bacteria which can lead to inhibitory or toxic effects if they are not acclimated to this environment [227,228]. The environmental stress causes the bacteria to go through plasmolysis and/or loss of cell activity, or to produce excess SMP and EPS which lead to membrane fouling, by blocking membrane pores [228,229]. Acclimation of bacteria to the harsh environment will reduce EPS and SMP formation, consequently lowering the blockage of the membrane [208]. Additionally, selecting a suitable type of bacteria would improve biodegradation efficiency in the MBR system when treating oily wastewater. Halotolerant microorganisms isolated from saline environments such as saltwater, sea mud, and a saline lake inoculated in MBR systems would eliminate the need for long acclimation periods to saline conditions in the bioreactor.

7. Conclusions

The discharge of oily wastewater leads to serious environmental problems if released without sufficient treatment due to the recalcitrant nature, toxicity, and carcinogenicity of the hydrocarbon compounds. The generation of the high volume of oily wastewater is an alarming threat to the ecosystem, and, therefore, numerous oily wastewater treatment approaches have been studied to achieve an eco-friendly method that will enhance treatment efficiency. In this regard, a comprehensive review has been conducted to provide a perspective regarding the practical aspects of HF membrane technology for oily wastewater treatment. Reviewing HF membrane filtration studies for the treatment of oily wastewater demonstrated that optimizing operational parameters and membrane modifications can significantly improve HF membrane performance in terms of maintaining membrane flux and reducing fouling. High SRT with high sludge age is desirable for oily wastewater treatment since it allows for biomass acclimation. Moderate aeration is also required for ideal membrane performance. Modified HF membranes with good hydration capability facilitate the passage of water and lead to high permeability, resulting in reduced membrane fouling.

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