EXPERIMENTAL INVESTIGATION OF A PILOT-SCALE MEMBRANE FILTRATION SYSTEM FOR OILY WASTEWATER TREATMENT

by

Parisa Keyvan Hosseini

Submitted in partial fulfillment of the requirements
for the degree of Master of Applied Science

at

Dalhousie University
Halifax, Nova Scotia
November 2022

I dedicate this thesis to my parents who taught me to be honest and real in all walks of life and pursue my dreams courageously. And to my husband for his unconditional love and devotion.

Table of Contents

List of Tables

List of Figures

Abstract

In this study, a pilot-scale submerged membrane filtration system using hollow fiber (HF) polytetrafluoroethylene (PTFE) membranes was experimentally investigated to treat synthetic oily seawater containing heavy crude oil (Cold Lake Dilbit (CLD)) and light crude oil (Very Low Sulfur Fuel Oil (VLSFO)). In this research project, the effect of different operating parameters such as aeration flow rate and membrane flux using different initial oil concentrations on the ultrafiltration (UF) membrane performance was examined. The total petroleum hydrocarbon (TPH) removal efficiency was found to be more than 91%. The results from the experiments met MARPOL 73/78 regulation which defines the oil content in the treated water does not exceed 15 ppm. Different fractions of petroleum and polycyclic aromatic hydrocarbon (PAH) compounds were reduced as well. The overall performance of this system in treatment of synthetic oily seawater proves its promising capability as an onsite treatment technology in oil spill response operations.

List of Abbreviations and Symbols Used

AOPs	Advanced Oxidation Processes
BTEX	Benzene, Toluene, Ethylbenzene, Xylene
CFV	Crossflow Velocity
CLD	Cold Lake Dilbit
COD	Chemical Oxygen Demand
CP	Concentration Polarization
DAF	Dissolved Air Floatation
HF	Hollow Fiber
MBRs	Membrane Bioreactors
MF	Microfiltration
MPRI	Multi-Partner Research Initiative
MSD	Mean Square Deviation
NF	Nanofiltration
PAC	PolyAluminum Chloride
PAHs	Polycyclic Aromatic Hydrocarbons
PHCs	Petroleum Hydrocarbons
PLC	Programmable Logic Controller
ppb	Parts Per Billion
ppm	Parts Per Million
PVDF	PolyVinyliDene Fluoride
PTFE	PolyTetraFluoroEthylene

RIS	Resistance-In-Series
RO	Reverse Osmosis
S/N	Signal to Noise Ratio
SS	Suspended Solid
TDS	Total Dissolved Solids
TMP	Transmembrane Pressure
TOC	Total Organic Carbon
TPH	Total Petroleum Hydrocarbon
UF	Ultrafiltration
UV-Vis spectrometry	Ultraviolet-Visible spectrometry
VOCs	Volatile Organic Compounds

Acknowledgements

At this juncture of my education at Dalhousie university, I have received support on the part of my supervisor who has helped me grow and learn everyday. I would like to express my deepest appreciation to Professor Lei Liu for his scientific insights, guidance, and support during this learning process.

I would also like to thank my supervisory committee, Professor Margaret Walsh and Professor Haibo Niu for their interest and assistance. Your encouragement, positive energy, and valuable comments are highly appreciated.

I gratefully acknowledge the financial support provided to this project by Department of Fisheries and Oceans Canada (DFO), Canada Coast Guard (CCG), Multi-Partner Research Initiative (MPRI) program, Natural Sciences and Engineering Research Council of Canada (NSERC), and the Faculty of Graduate Studies for FGS scholarship.

This endeavor would not have been possible without the support of Dr. Kenneth Lee whose inspirational role has guided me to move forward. Being a member of MPRI has been my great pleasure and an unforgettable experience.

The big portion of my educational journey at Dalhousie was coincided with COVID-19 pandemic. During hard times of pandemic, the assistance and companionship of Dr. Hesam Hafezi and Dr. Naznin Sultana Daisy have been always perpetual in my mind and heart.

I would like to acknowledge my fellow graduate students whose friendship throughout this journey has proven to be invaluable.

I would also like to extend my appreciation to civil and chemical engineering technicians Jesse Keane, Heather Daurie, Danniel Chevalier, and Gerald Fraser for all their assistance along the way. In addition, I am grateful to Professor Navid Bahrani for his helps during the experimental runs.

Words cannot express my gratitude toward my parents whose selfless love, care, and sacrifice made me the person who I am. Big thanks should go to my twin sister, Mahsa, who has been always with me during this journey, and we experienced lots of laughs and cries with each other. I owe thanks to a very special person, my husband Pouria, whose unfailing love and understanding have been precious throughout my educational journey. Also, I express my thanks to my brother, Parsa, you are awesome.

CHAPTER 1 INTRODUCTION

This chapter contains the background which explains the problem statement and motivation for this research, determines the research purposes, and outlines the thesis layout.

1.1. Background

The increase in energy demand and economic growth has resulted in continuous gas and oil exploration and production which significantly increases the risk of oily wastewater discharge into the environment (Feng et al., 2021). One major part of releasing oily wastewater into the ecosystem is associated with marine oil spill incidents. For instance, 140 significant oil spill events have released more than 7 million tonnes of oil into the environment throughout the last century (Fingas, 2016). Some major oil spill events such as SeaRose (250 m^3 of spilled oil, Newfoundland and Labrador, Canada, 2018), Deepwater Horizon (500,000 m^3 of spilled oil, Gulf of Mexico, 2010), Montara (23,500 m^3 of spilled oil, Western Australia, 2009), Hebei Spirit (30,865 m^3 of spilled oil, South Korea, 2007), Prestige (48,138 m^3 of spilled oil, Spain, 2002), Exxon Valdez (41,000 m^3 of spilled oil, Alaska, USA, 1989), and Odyssey (176,300 m^3 of spilled oil, Nova Scotia, Canada, 1988) adversely affected marine environment, ecosystems, and human health (Nerubenko, 2015; Bonisoli-Alquati et al., 2016; Fingas, 2016; Spies et al., 2017; Yim et al., 2017; CBC, 2019; Galieriková & Materna, 2020; CBC, 2021). As an example, the Deepwater Horizon oil spill is officially the largest oil spill in history, which led to a widespread contamination of coastal habitats and ecosystems, and negatively affected fisheries, tourism, and other societal sectors (Beyer et al., 2016). The harmful impacts of other oil spill disasters on the environment, economy, and public health have also been documented (Penela-Arenaz et al., 2009; Gill et al., 2012; Young et al., 2011; Dale et al., 2013; Morales-Caselles et al., 2017; Yim et al., 2017; Avery, 2020).

In marine oil spill response operations, different techniques such as manual recovery, booming, skimming, and sorption as physical/mechanical treatment methods are used to minimize or avoid the negative impacts of oil spills. In addition, chemical treatment methods such as in-situ burning, dispersion, solidification, and demulsification; and natural reduction and bioremediation as biological treatment methods are utilized in marine oil spill cleanup operations (Li et al., 2016).

Currently, oleophilic skimmers collect on average 30% oil and 70% water. The large volume of water collected is unavoidable due to wave conditions in the ocean that contributes to variations in oil slick thickness, shearing of slick surface, and mixing of oil with underlying water in a spill region, which affect the performance of skimmers (Chen et al., 2022). The efficiency of a skimmer is also influenced by the viscosity of the spilled oil. In general, lighter viscosity crude oils do not easily accumulate in thick layers on the surface of oleophilic skimmers; therefore, more water is collected. High viscosity oils are excessively sticky and are difficult to remove from the surface of the skimmer; thus, once the skimmer is saturated with oil, it starts to collect a large volume of water. Oleophilic skimmers are most efficient at collecting medium viscosity crude oil (ITOPF, 2012). As a result of the inefficiencies of the skimmers, surplus water is recovered from the operation and placed in temporary storage on the vessels. To obtain optimal efficiency for oil collection, the skimmers would have to be dynamically adjusted throughout the operation, which is impractical (Chen et al., 2022).

The present oil spill response practices require that all fluids collected during the operation are transported back to shore. Therefore, most of the storage space is taken up by water, which increases the requirement for additional temporary storage on the barges and/or the need for additional trips to transport the recovered fluids back to shore for treatment and disposal. The response capacity and efficacy of oily wastewater treatment is significantly restricted by current oil spill response policies, allowing for further spill dispersion and increase of environmental harm (Chen et al., 2022). Natural oil decantation (i.e., gravity separation) in the temporary storage tanks occurs during the transportation of recovered fluids back to shore. The fluids separate into layers according to their densities, with free floating oil at the top, and water at the bottom (Ghidossi et al., 2009), where water takes up 50 to 66% of the total storage space. The disposal of decanted water at sea is usually regulated by an acceptable overall oil content. For example, the International Spill Control Organization (ISCO) suggests that the discharge of decanted water follow the regulation of International Convention for the Prevention of Pollution from Ships (MARPOL 73/78), which states that the allowed oil content in the discharged water must be less than 15 ppm (ISCO, 2022). If the decanted water can be treated to meet the MARPOL 73/78 regulation, then the treated water can be discharged into the aquatic environment freeing up the majority of the storage space for further oily wastewater treatment. This will improve the capacity and efficiency of the oil spill response operations and will not harm the environment (Chen et al., 2022).

Membrane filtration technology has been proposed as an effective onsite treatment method for oily wastewater generated from marine oil spills (Han et al., 2019). Membranes are semi-permeable barriers, through which selectivity between species is achieved to allow for the separation between unwanted and wanted particles. Therefore, desired species pass through the membrane while undesirable ones are retained in the bulk solution (Kang et al., 2019). Membrane filtration systems have proven to have a high hydrocarbon removal capacity, simple operation, small footprint, low chemical usage, and low secondary waste production (Singh et al., 2011; Tanudjaja et al., 2019; Sharghi et al., 2020). Among different membrane configurations, hollow fiber (HF) membranes are used for treatment of complex wastewaters (i.e., oily wastewater) (Kose et al., 2012; Razavi & Miri, 2015; Johari et al., 2020; Ayub et al., 2021). HF membranes are preferred over other membrane configurations because they are compact modules with very high membrane surface area, high packing density, feasibility of backwashing, self-supporting structure, simplicity of handling, and low manufacturing cost (Baker, 2012; Wan et al., 2017; Akhondi et al., 2017; Obotey Ezugbe & Rathilal, 2020). Although membrane-based technologies are effective treatment systems, membrane fouling is a challenging issue hindering its large-scale applications (Pendashteh et al., 2011; Bagheri & Mirbagheri, 2018). Membrane fouling refers to the accumulation of substances on the membrane surface or within the pores which results in increasing transmembrane pressure (TMP), decreasing permeate production, and membrane lifespan, and consequently frequent membrane cleaning and replacement are required (Bagheri & Mirbagheri, 2018; Huang et al., 2018).

Reviewing previous studies demonstrates that the majority of the research into the use of membrane filtration for oily wastewater treatment has been conducted at bench-scale, and most of the laboratory experiments were performed under the constant TMP. Constant TMP is not appropriate for larger-scale systems because solute particles move towards the membrane surface more quickly than they are transported away. This results in accumulating more particles on the membrane surface at constant TMP than at constant flux condition; therefore leading to more rapid and serious membrane fouling (Sentana et al., 2009; Hussain, 2019; Keyvan Hosseini et al., 2023). Limited published studies have evaluated hollow fiber (HF) membrane systems to treat oily wastewater, with most studies having evaluated flat-sheet membranes (Li et al., 2017; Hube et al., 2020). Flat-sheet membranes are fragile and have low packing density and low membrane flux (Doyen et al., 2010; Obotey Ezugbe & Rathilal, 2020; Bopape et al., 2021). These membranes are prone to rapid and frequent fouling due to their restricted backwashing capability (Altinbas et al., 2021). HF membranes are more beneficial in comparison with flat-sheet membranes as a result of their higher membrane flux due to higher packing density and reduced membrane fouling as a consequence of aeration (Akhondi et al., 2017; Wan et al., 2017; Altinbas et al., 2021; Keyvan Hosseini et al., 2023).

In addition, most of the membrane filtration research conducted to date has evaluated a side-stream configuration, which refers to the condition when membrane is placed outside the membrane tank. In this configuration, serious membrane fouling occurs, and thus, frequent membrane cleaning is necessary (Judd & Turan, 2018; Obotey Ezugbe & Rathilal, 2020; Tran et al., 2021). In the submerged type, the membrane is located inside the membrane tank and the bubbles scouring is used as a common practice to remove cake layer from the membrane surface. Other techniques such as mechanical vibrations and particle fluidization are employed to decrease membrane fouling; therefore, this type has lower membrane fouling and more efficient membrane performance (Wang et al., 2009; Lin et al., 2012; Judd & Turan, 2018; Tran et al., 2021; Keyvan Hosseini et al., 2023).

One of the main concerns for the discharge of decanted water into the ocean is the toxicity of petroleum hydrocarbons (PHCs) in the decanted water and its potential effects on marine environment and ecosystems (Perhar & Arhonditsis, 2014). PHCs and PAHs are comprised of mutagenic and carcinogenic components and can bioaccumulate in human and animal tissues (Hodson, 2017). Most of the membrane filtration studies have investigated the total petroleum hydrocarbon (TPH) removal efficiency, and there have been no membrane filtration experiments that examine the removal efficiency of polycyclic aromatic hydrocarbons (PAHs) and other specific petroleum fractions from oily wastewater (González-Pérez et al., 2012; Kose et al., 2012; Syafiuddin & Boopathy, 2021; Keyvan Hosseini et al., 2023).

To address these knowledge gaps, the purpose of this research project was to investigate the performance of a pilot-scale membrane filtration system equipped with two polytetrafluoroethylene (PTFE) HF membranes for the treatment of synthetic oily seawater containing heavy and light crude oil using ultrafiltration (UF) membrane under the constant flux. The performance of the membrane filtration system was studied in terms of total hydrocarbon removal efficiency, different petroleum fractions (F1-F4), particular PAH compounds removal efficiency, and membrane fouling. PTFE membrane material was chosen for this research because of its high chemical resistance, mechanical strength for backwashing the membrane at high flow rates which reduces membrane fouling, and antifouling features that increase the membrane lifespan (Song et al., 2017; Li et al., 2018).

1.2. Research Objectives

The overall objective of this research was to examine the capability of a membrane filtration system to treat synthetic oily seawater. The specific objectives of this thesis were as follows:

- Prepare a stable oil-in-water emulsion using a high-shear vertical mixer
- Determine the effect of operating parameters (i.e., aeration flow rate and membrane flux) using different initial oil concentrations on the performance of a pilot-scale membrane filtration system equipped with PTFE-UF-HF membranes in terms of hydrocarbon removal efficiency.
- Investigate the impact of membrane flux and aeration flow rate on membrane fouling during the treatment of synthetic oily seawater comprised of heavy or light crude oil.

1.3. Thesis Outline

This thesis is divided into five chapters. Figure 1.1 shows thesis organization for each chapter.

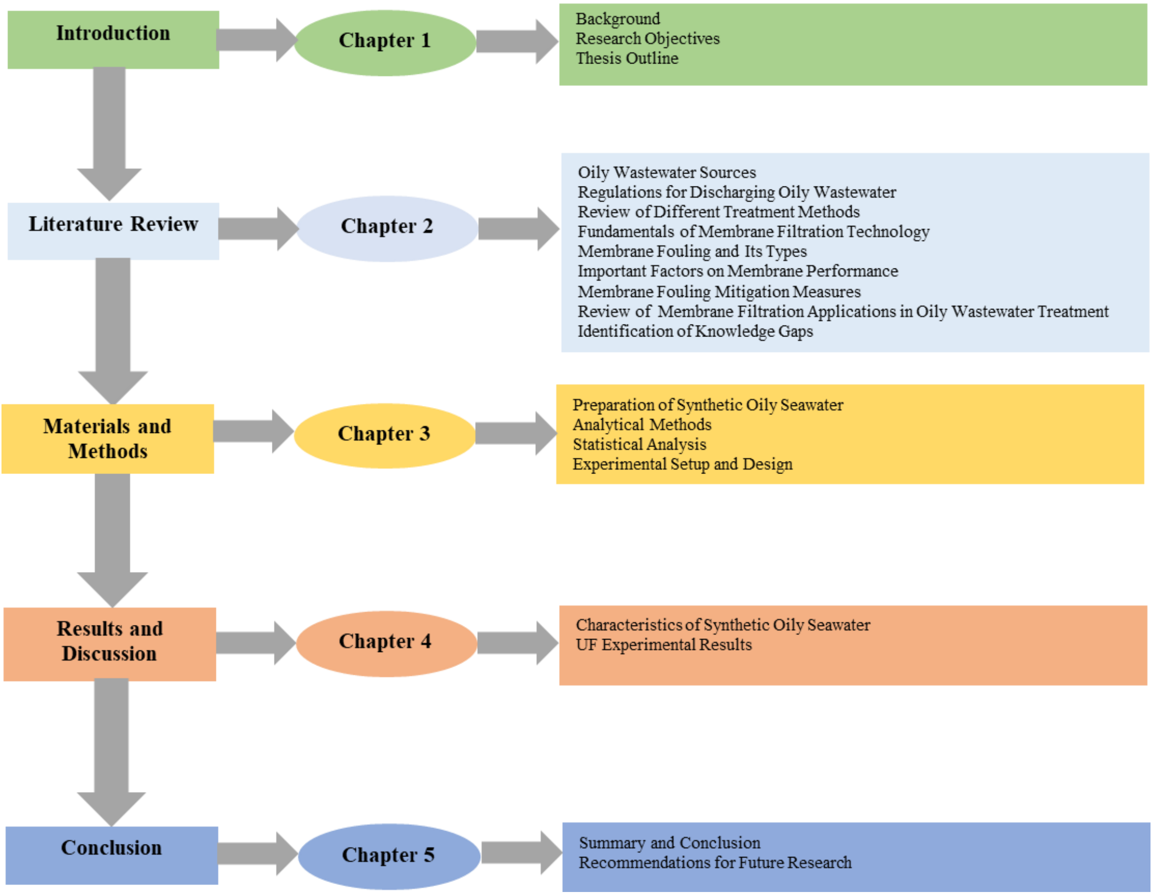

Figure 1.1 Chart of thesis organization

Chapter 1 introduces the project background, problem statement, and the necessity for this research, in addition to defining the research objectives, and thesis organization.

Chapter 2 provides a literature review associated with the application of membrane filtration systems in the treatment of oily wastewater to better guide the readers throughout this research and help identify research gaps. The identification of industrial oily wastewater sources and their environmental impacts, regulations for discharging oily wastewater, different oily wastewater treatment methods, fundamentals of membrane filtration system, membrane fouling and its types, significant factors affecting membrane performance, and membrane fouling mitigation measures are provided as well.

Chapter 3 outlines the materials and methods used to obtain the research objectives. The research plan was classified into four primary steps in order to accurately achieve this goal. The steps included weathering and emulsifying crude oils to create a synthetic oily wastewater to use in experiments, running the pilot-scale PTFE-UF-HF membrane filtration system and adjusting the operating parameters, sampling, and analyzing different parameters to understand hydrocarbon removal as well as investigating membrane fouling.

Chapter 4 provides the results and discussion through different analyses during the membrane filtration process in the treatment of synthetic oily seawater. Chapter 5 summarizes the main findings and conclusions of the study and recognizes areas which may need further research. Sources and references are provided in the last section of this thesis.

CHAPTER 2 LITERATURE REVIEW

This chapter begins with reviewing different sources of industrial oily wastewater and its environmental impacts, regulations for discharging oily wastewater, and various treatment methods. Second, membrane filtration technology in terms of membrane features, module configurations, membrane fouling and its types is discussed. Third, factors affecting the performance of membrane filtration systems, and membrane fouling mitigation methods are considered. Fourth, the application of membrane filtration technology for treatment of oily wastewater is investigated. Finally, the knowledge gaps associated with membrane filtration systems in treatment of oily wastewater are stated to highlight the research requirement.

2.1. Oily Wastewater Sources

Different industries such as petroleum refineries, metallurgy, and transportation have mainly contributed to the large production of oily wastewater (Kuyukina et al., 2020). Oilfield wastewater is mostly comprised of produced water which is defined as any water in a reservoir with a hydrocarbon resource and produced with crude oil or gas (Mondal & Wickramasinghe, 2008; Soltani et al., 2010). The total amount of produced water globally generated from the petroleum industry is 39.746×10^9 L/d (Sanghamitra et al., 2021). In various sites, the volume of water is 2-5 or up to 50 times more than the volume of oil produced (Veil, 2015). The rate of oil extraction, extraction technology, and reservoir characteristics determine the volume of produced water (Ahmad et al., 2005; Bakke et al., 2013; Veil, 2015). The generated wastewater is comprised of benzene, toluene, ethylbenzene, and xylene (BTEX), PAHs, phenols, surfactants, biocides as toxic organic components and mineral salt, heavy metals, and radioactive elements as toxic inorganic compounds which are environmental concerns (Kuyukina et al., 2020).

Metal processing industries are another large source of oily wastewater production; the generated oily wastewater can be highly viscous, limiting the capacities of wastewater treatment plants, and resulting in higher maintenance costs and energy usage (Yang, 2007; Kuyukina et al., 2020). Globally, more than 2,000,000 m^3 of metal processing fluid are used annually; however, the volume of wastewater can be ten times higher because of the dilution of this wastewater. Emulsified oil, emulsifiers, surfactants, suspended particles, and metals are the main constituents of metallurgical oily wastewater (Wu et al., 2017).

Another major source of oily wastewater is the transportation industry; one big portion of this source is associated with the shipping industry leading to illegal discharge of wastewater generated from ships (i.e., shipboard slop wastewater, bilge and ballast water), and oil spill disasters (Han et al., 2019). For instance, shipboard slop wastewater is produced through washing oil tankers with seawater and removing hydrocarbons from the walls of the tankers. It has high salinity and hydrocarbons, and variable organic contaminants which may impact the treatment of specific slop wastewater (Campo et al., 2017). Ballast and bilge waters are other types of wastewaters from ships with toxic compounds which are harmful because of petroleum and petroleum-derived products that have carcinogenic properties (Karakulski & Gryta, 2017). Their salinity may be different from vessel to vessel and may also vary daily on the same vessel. Typical salinity concentrations are between 0 and 15 g/L (Sun et al., 2010).

Over the years, oil exploration and production activities have increased, and millions of tonnes of petroleum-based products are transported through open waterways worldwide, increasing the risk of oil spills (Abidli et al., 2020). There are three categories of oil following oil spill incidents: the first classification is light oil which is moderately volatile; their residue remains after a few days; light oils include diesel, heating oil, and light crudes. They are not accumulated in large quantities on banks and shorelines. The second category is medium crude oils, which evaporate moderately within 24 h; most crude oils belong to this category. Lastly, depending on the ocean and weather conditions, heavy oils cannot be quickly evaporated and dispersed in the sea at a specific rate. They become dense, and form sticky materials called tar balls and asphalt. These substances are difficult to eliminate from rocks and sediments. When they are deposited, more aggressive cleanup operations are necessary compared to lighter ones. Bitumen, Dilbit, refined fuel oils such as No. 6 fuel oil, and Bunker C are heavy oils (USEPA, 1999; Lee et al., 2015).

2.2. Oily Wastewater Characteristics and Its Environmental Impacts

Oily wastewater is classified based on droplet size in the dispersed phase; droplets larger than 150 µm are considered as free floating, while droplets between 20 and 150 µm are designated as dispersed, and emulsified oil has a droplet size of smaller than 20 µm. A droplet size of less than 5 µm is associated with dissolved oil (Medeiros et al., 2022). Oily wastewater contains volatile organic compounds (VOCs), PAHs, and other harmful and toxic compounds that impact the environment, wildlife, and human health. For instance, as a result of oily wastewater discharge, the physico-chemical qualities of soil are unfavorably affected by changing morphology and reducing hydraulic conductivity, which restrict seed germination, plant growth, and crop production (Adetunji & Olaniran, 2021). When oily wastewater is spilled into the marine environment, various marine species are negatively affected in terms of physiology and ecology, leading to habitat alteration, feeding destruction, and mortality (Visvikis & Panayides, 2017; Mohammadi et al., 2020). Carcinogens and contaminants can also be transferred to the human food chain, significantly harming human health (Cho, 2007). Chronic exposure to petrochemical hydrocarbons such as BTEX results in fatal leukemia and central nervous system dysfunction (Souza et al., 2014). It also affects the socioeconomic aspects of human life, such as aquaculture, fisheries, tourism, and local businessmen (Fingas & Brown, 1997; Dalaklis et al., 2020).

2.3. Regulations for Discharging Oily Wastewater

The discharge of oily wastewater into the environment with no appropriate treatment has adverse effects on the environment, wildlife, and human life; therefore, it is required to be cautiously regulated (Ipieca, 2013; Yu et al., 2017). To deal with this issue, different region-specific regulations have been enacted. For instance, the Canada Fisheries Act (Government of Canada, 2018) prohibits the discharge of decanted oily wastewater into the ocean. Under MARPOL 73/78 regulation (ISCO, 2022), the release of oil by vessels into the sea is not permitted, with the following exemptions: the oil content of the discharge does not exceed 15 ppm (except in Special Areas, such as the Antarctic or the Great Lakes, where no oil content is permitted) (SL Ross, 2005). The United States Environmental Protection Agency (USEPA, 2016) specifies the oil and grease discharge limit of 29 mg/L for monthly average with a maximum daily discharge of 42 mg/L (Souza et al., 2020). In the North Sea region, Oslo-Paris (OSPAR) convention (OSPAR commission, 2015) legalizes that the upper limit for the discharge of oil content in the wastewater is 30 mg/L (Dickhout et al., 2017). In Norway, the allowable amount of oil concentration to be discharged into the sea is 30 mg/L (Yu et al., 2017). In order to meet these regulations, technologies should be able to treat oily wastewater and reduce the oil concentration to reach the treatment goals.

2.4. Treatment Methods for Oily Wastewater

Different technologies have been utilized to treat oily wastewater which are selected proportional to the source of wastewater, the diversity and levels of contaminants, and the subsequent intended use of treated effluents (Kuyukina et al., 2020). Physical treatment methods are used to remove free oil and grease fractions and suspended particles (Wei et al., 2019). Gravity separation, dissolved air flotation (DAF), and membrane filtration are categorized as physical methods to remove oil from oily wastewater. Gravity separation operates based on density difference of oil and water (Le et al., 2013). This technique is straightforward but it has some drawbacks such as long-time operation, large footprint, and low separation efficiency particularly for separating dispersed and emulsified oils (Medeiros et al., 2022). Therefore, it is mainly used as a pre-treatment or primary treatment (Medeiros et al., 2022). Conventional DAF produces micro-sized bubbles which attach to the oil droplets and raise the buoyancy of droplets to move them to the water surface and remove dispersed and emulsified oils. This technology has a small footprint and fast operation but requires high energy input to producing micro-sized bubbles (Le et al., 2013; Padaki et al., 2015). To overcome the drawbacks of two previous methods such as prolonged process, large-space requirement, high energy consumption, and low treatment efficiency, membrane filtration has been proposed. In this technology, membrane pore size acts as a selective barrier allowing the smaller particles to pass through, whereas large oil particles are physically separated from the water phase through sieving and maintained in the feed solution (Goh et al., 2019; Ahamed & Lichtfouse, 2021). This technology has been shown to be a viable option for treating oily wastewaters, particularly for effluents containing emulsified oil (Tanudjaja et al., 2019; Sharghi et al., 2020).

2.5. Membrane Filtration Technology

In 1755, Abbé Nollet introduced water permeation by a thin sheet of animal bladders in France (Sempere, 2015). Following that, Dr. Adolf Eugen Fick from Germany introduced diffusion law and the first high-pressure synthetic membrane made of nitrocellulose (Ahmed et al., 2017). In 1855, Fick was the first person who reported the oldest membrane which was MF membrane made of cellulose nitrate (Anis et al., 2019). In 1877, the enhancement of membrane performance was initially conducted through surface modification by W. Pfeffer. He created a copper ferrocyanide layer on a porous porcelain membrane to study osmosis. He modified the membrane by saturating the porous porcelain with a copper sulfate solution, followed by placing one of the faces of the membrane into a potassium ferrocyanide solution. This membrane was used for studying the osmotic flow of water from a pure water reservoir to a saline water reservoir. Surface modification of membranes sustained into the 20th century as explained in a sequence of papers published in 1907 and 1908 (Miller et al., 2017).

In 1907, Dr. Bechhold coined the term “ultrafiltration” for his collodion membranes (Bechhold, 1907). These low-pressure cellulosic membranes were prepared by impregnating filter paper by glacial acetic acid and gelatin. Following Bechhold’s membrane, there have been continuous attempts to enhance UF membranes. Zsigmondy, Bachman, and Ferry further developed Bechhold’s membrane fabrication method (Ahmed et al., 2017). Microporous cellulosic membranes including cellulose nitrate, and cellulose di/triacetate were commercially available by the early 1930s (Elford et al., 1935; Baker, 2004). During the 1940s, the number of membrane manufacturers grew but until the mid-1960s, MF membranes were only used in laboratories and very small-scale industries (Gul et al., 2021). Around the 1950s, many industrial firms in the US were founded that started using membrane technology for separation procedures and other large-scale applications, this drove the search for advances in this technology (Potts et al., 1981). The first trials for treatment of oily wastewater using membrane separation processes date back to the early 1970s (Zare et al., 2013). Since then, different companies have joined to manufacture various types of membranes and applied them in treating oily wastewaters. Manufactured membranes from companies such as Zenon, Kubota, Koch, Sumitomo, and Toray have been used in the treatment of oily wastewater which is one of the most complicated industrial wastewaters because of oil nature, its forms in water, its content, and concentration (Zhidong et al., 2009; Buer & Cumin, 2010). Membrane filtration has been successful in the treatment of oily wastewaters, particularly for the effluents containing emulsified and dissolved oils (Han et al., 2015; Li et al., 2017).

2.6. Membrane Property and Module Configuration

Membrane as a barrier has been an efficient and reliable way to selectively transport certain components based on their characteristics (Kang et al., 2019). Membranes are very diverse in nature, and their properties rely heavily on the type of the materials used, such as ceramic and polymeric materials. Ceramic membranes have a high hydrophilicity, membrane fouling resistance, integrity, and chemical resistance which can generally lead to a high filtration performance. However, high fabrication cost makes them economically unfeasible. Polymeric materials have a high physical and chemical resistance, which has made them become the most typical and available materials for membrane fabrication with different pore sizes. Examples of polymeric materials include polyvinylidene fluoride (PVDF), polyethersulfone (PES), polysulfone (PS), polyethylene (PE), polypropylene (PP), and polytetrafluoroethylene (PTFE). Since most of the polymeric materials are hydrophobic, hydrophilic technology has been developed to modify the membrane surface with hydrophilic polymers to make the membrane hydrophilic. The resulted hydrophilicity could not only help prevent the oil droplets blockage on the membrane surface and improve the treatment efficiency, but also help save a significant amount of cost in membrane maintenance and replacement (Mutamim et al., 2012; Judd, 2016; Cheng et al., 2017; Melbiah et al., 2017). When a membrane is in a direct contact with oily wastewater, the membrane materials should be robust enough for a long period of time and sustain a good performance according to its chemical resistance, mechanical strength, durability in wide range of pH levels, hydrophilicity, and membrane flux (Abdel-Karim et al., 2017).

Membrane materials can be manufactured in four main membrane configuration categories, such as plate and frame, tubular, spiral wound, and HF which are compared in Table 2.1 (Marrot et al., 2004; Judd, 2016).

Table 2.1 Comparison of different membrane configurations (Keyvan Hosseini et al., 2022)

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 backflushable, the lowest membrane area per unit volume, low efficiency compared to other configurations, high pressure drop	Hybrid MF/UF: 99.9% UF: >95%	(Frederickson, 2005; Judd, 2010; Masoudnia et al., 2014; Huang et al., 2015; Berk, 2018; Obotey Ezugbe & Rathilal, 2020)
Tubular Membrane Module	MF/UF, wastewaters 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 backflushable, very high cost, very large footprint	UF: 99% UF: 98.04%	(Kim et al., 1998; Frederickson, 2005; Li et al., 2006; Judd, 2010; Obotey Ezugbe & Rathilal, 2020)
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 backflushable	UF: 90.1% UF: 99.7%	(Cheryan & Rajagopalan, 1998; Marchese et al., 2000; Schwinge et al., 2004; Radjenovic et al., 2008; Judd, 2010; Obotey Ezugbe & Rathilal, 2020)

HF Membrane Module	MF/UF and RO, MBR, industrial wastewater, oily wastewater juice processing, biotech applications	Moderate capital cost, very high packing density, backflushable, 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%	(Frederickson, 2005; Radjenovic et al., 2008; Judd, 2010; Kose et al., 2012; Salahi et al., 2015; Obotey Ezugbe & Rathilal, 2020)

Plate and frame configuration has a restricted application and mostly used for treating wastewater with high amounts of suspended solids (SS) (Obotey Ezugbe & Rathilal, 2020). Its manufacturing cost is high, and the packing density is in the range of 148 to 492 m^2/m^3 which is lower than spiral wound and HF. This membrane has a moderate potential of membrane fouling, and its cleaning is easier than spiral wound and HF. In tubular membrane, the manufacturing cost is high, and its packing density is 20 to 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 (Berk, 2018; Obotey Ezugbe & Rathilal, 2020; Keyvan Hosseini et al., 2022). This membrane has a low potential of membrane fouling, and its cleaning is the easiest due to the large diameter (Berk, 2018). In spiral wound, packing density is 492 to 1247 m^2/m^3 which is lower than HF. This membrane has a high potential of membrane fouling, is hard to clean, and has a moderate manufacturing cost (Obotey Ezugbe & Rathilal, 2020; Keyvan Hosseini et al., 2022). The packing density of HF is 492 to 4924 m^2/m^3 which is the highest and the manufacturing cost is low. Generally, HF membranes are favored over other membrane configurations due to their benefits such as compact modules with very high membrane surface area per unit volume, high packing density, and capable of generating movements by mechanisms (i.e., bubbling) to mitigate membrane fouling (Di Profio et al., 2011; Baker, 2012; Keyvan Hosseini et al., 2022).

Based on the difference between the pressure-driven membrane processes, membranes are classified into four groups such as MF, nanofiltration (NF), UF, and reverse osmosis (RO). The four categories are given by the differential pressure applied to speed of the mass transport through the membrane. In this line, higher pressure is needed to separate smaller components according to their geometrical dimension (Barjoveanu & Teodosiu, 2006; Padaki et al., 2015; Martini et al., 2017; Varjani et al., 2020). MF membranes are used for removing suspended solids with the particle size in the range of 0.1-5 µm. UF and NF are used for removing particles in the range of 0.001-0.1 µm and 0.0001-0.01 µm, respectively (Le-Clech et al., 2006). The particle dimension for RO ranges between 0.0001 to 0.001 µm and this membrane is able to remove salinity from wastewater (Jamaly et al., 2014). MF and UF membranes have been widely used for treatment of oily wastewater compared to NF and RO. This is because of their capability in removing oil particles due to their selectivity features and separation mechanism. In both MF and UF membranes, the removal mechanism is sieving, and transport mechanism is convection. NF and RO are broadly utilized to remove salts and their removal and transport mechanisms are diffusion (Barjoveanu & Teodosiu, 2006; Cao, 2016; Abuhasel et al., 2021; Keyvan Hosseini et al., 2022).

Membrane filtration systems have two main types of process flow configurations (shown in Figure 2.1) based on the arrangement of the membrane modules: side-stream and submerged.

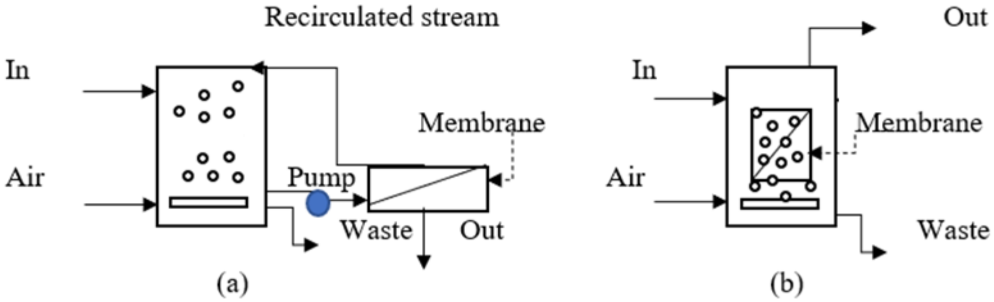

Figure 2.1 a) side-stream b) submerged (Modified figure from Judd, 2010)

Side-stream configuration refers to when the membrane module is outside of the membrane tank while submerged type refers to the configuration which the membrane module is directly placed inside the membrane tank (Le-Clech et al., 2005). For small flows of effluents, the side-stream membrane filtration system is often selected because it is simpler in operation and smaller in footprint (Marrot et al., 2004; Buer & Cumin, 2010). In side-stream configuration, the mixed liquor from the reactor is circulated across the membrane at high crossflow velocities and pressures to mitigate membrane fouling (Kharraz et al., 2022). This configuration requires high energy for circulation, and has severe membrane fouling (Park et al., 2018; Al-Khafaji et al., 2022). For very large plants, the submerged membrane filtration system is mostly selected due to its lower tangential velocities which results in milder operating conditions (Lin et al., 2012). This configuration has also lower energy consumption, and overall robustness in performance compared to side-stream systems (Marrot et al., 2004; Buer & Cumin, 2010; Judd, 2016). The cleaning process in submerged configuration is more complicated than side-stream type. Increasing aeration flow rate, decreasing the membrane flux, and using physical or chemical cleaning are three methods to manage membrane fouling in this configuration. Aeration generates a shear force on the membrane surface through the rise of bubbles which decreases the accumulation of foulants on the membrane surface. Using lower membrane flux reduces membrane fouling since it diminishes the rate of foulants reaching the membrane surface. These methods can significantly mitigate membrane fouling and the requirement for membrane cleaning. In addition, physical cleaning is preferred over chemical cleaning due to its simplicity and harmless material usage which does not generate chemical waste (Judd, 2005; Al-Khafaji et al., 2022; Keyvan Hosseini et al., 2022).

2.7. Membrane Fouling

One of the most challenging issues in membrane filtration technology is membrane fouling which refers to the accumulation and deposition of substances on the surface of the membrane due to complex physical and chemical interactions among various foulants (Li & Elimelech, 2004; Gkotsis et al., 2014; Krzeminski et al., 2017). In oily wastewater, oil interacts with a membrane surface in distinctive ways: they deform, coalesce, break up, enter, and seal membrane pores. Changes in oil shape allows oil droplets to enter the membrane pores and can permeate across the pores smaller than the droplet size (Tummons et al., 2017, 2020). This results in flux decline over time when TMP is constant and increase in TMP over time when membrane flux is constant. Fouling phenomenon exacerbates the membrane performance, decreases time intervals for membrane cleaning and replacement, and shortens membrane lifespan leading to increase in operational cost (Safazadeh Haghighi, 2011).

2.8. Types of Membrane Fouling

There are different major classes of membrane fouling based on the location of fouling, and intensity and flux recovery. Depending on the fouling location, fouling can be classified as concentration polarization (CP), external fouling, or internal fouling (Bagheri & Mirbagheri, 2018). In CP, solutes and ions are accumulated in the thin liquid layer close to the surface of the membrane (Bhattacharjee et al., 1999; Li et al., 2016). This will lead to the formation of a region with higher concentration compared to the whole bulk (Romero & Davis, 1991). It increases flow resistance and decreases membrane flux (Abdelrasoul, 2015). CP is a reversible phenomenon and does not impact the intrinsic properties of the membrane (Shi et al., 2014).

In addition to CP, particles, colloids, and macromolecules increase membrane resistance through adsorption or deposition on the surface of the membrane which is called external fouling. This type of fouling creates a layer on the membrane surface categorized as gel layer and cake layer. Due to the difference in pressure between the feed and permeate sides of the membrane, macromolecules, colloids, and inorganic solutes are deposited on the surface of the membrane and form a gel layer. Cake layer is created with the accumulation of solids on the membrane surface (Blandin et al., 2016). Adsorption and deposition of solutes and colloidal particles within the pore refers to the internal fouling or pore blocking (Stephenson et al., 2000).

Based on flux recovery and degree of foulants removal, membrane fouling can also be classified as reversible fouling, residual fouling, irreversible fouling, or irrecoverable/permanent fouling (Gkotsis et al., 2014). Reversible fouling can happen within 10 minutes of filtration and foulants can be removed by physical cleaning methods (i.e., relaxation and backwash). If foulants need maintenance cleaning such as chemically enhanced backflush, the fouling refers to residual fouling which occurs within 1 to 2 weeks. When the foulants cannot be removed by physical cleaning and need chemical cleaning, the irreversible fouling happens in the system typically within 6 to 12 months. Irrecoverable/permanent fouling takes place when the foulants cannot be removed by chemical cleaning which happens within several years (Kraume et al., 2009; Gkotsis et al., 2014; Bagheri & Mirbagheri, 2018; Du et al., 2020).

2.9. Factors Affecting Membrane Performance

2.9.1. Membrane Features

2.9.1.1 Pore Size Distribution

The pores of membrane should be small enough to provide an excellent size-sieving effect which will avoid pore clogging (Huang et al., 2018). For example, in membrane filtration systems for treatment of oily wastewater, small membrane pore size intensifies the pressure required for entering oil films and droplets into pores and this will decrease membrane fouling by emulsified oil (Tummons et al., 2020). However, if pores are too small, the membrane resistance to permeate flux will be too high which is not desired (Huang et al., 2018). Therefore, the size of the membrane pores should be carefully selected.

2.9.1.2 Surface Chemistry

Contact angle is a factor showing the wettability of a solid surface by a liquid which is formed by a liquid and the tangent line to the upper surface where three phase boundaries such as liquid, gas, and solid are interconnected (shown in Figure 2.2).

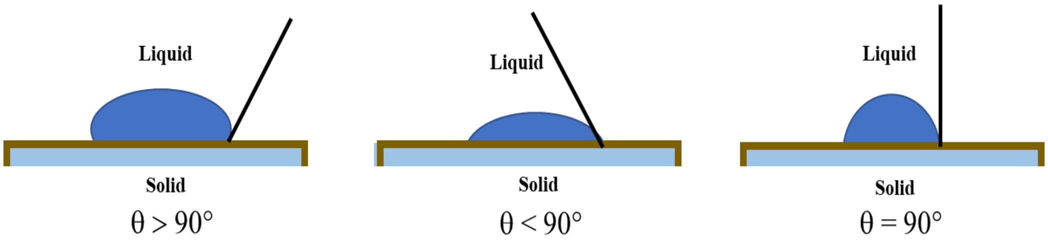

Figure 2.2 Demonstration of contact angle of a liquid (Modified figure from Safazadeh Haghighi, 2011)

The contact angle between a droplet and a solid surface demonstrates hydrophobicity or hydrophilicity. When the contact angle of a water droplet on the membrane surface is more than 90˚, the membrane surface is hydrophobic and when it is between 0˚ and 90˚, the membrane surface is hydrophilic (Safazadeh Haghighi, 2011). In oily wastewater treatment using membrane filtration system, oil droplet makes the membrane wet, which is distributed throughout the membrane surface, and water cannot be dispersed over the membrane surface. Therefore, instead of considering water droplets on the membrane surface, oil droplets are determined to be on the solid surface. Consequently, the condition of contact angle is reversed and if $\theta > 90^\circ$, the membrane is hydrophilic and when $0^\circ < \theta < 90^\circ$, the membrane is hydrophobic (Safazadeh Haghighi, 2011; Tummons et al., 2020). The affinity between oil droplets and the membrane under water such as underwater oleophilicity or olephobicity is a significant factor. In the membranes with an antifouling feature, affinity between oil and membrane under water is negligible and is called underwater oleophobicity. Membrane with higher hydrophilicity shows higher underwater oleophobicity; this causes hydrophilic membranes to be chosen for constructing antifouling membranes (Huang et al., 2018). If the oil droplets and membrane have different surface charges, membrane fouling will be exacerbated due to the electrostatic attraction (Lu et al., 2016). It is understood that in hydrophilic membranes, irreversible fouling is less and when the charge of membrane surface is similar to surfactant charge, membrane fouling will decrease (Dickhout et al., 2018; Huang et al., 2018; Saini et al., 2019). Therefore, membrane fouling can be impacted by membrane surface charges through modifying its wettability towards oil droplets (Shi et al., 2016; Liu et al., 2017).

2.9.2. Effect of TMP, Membrane Flux, and Crossflow Velocity (CFV)

Membrane performance is primarily determined by TMP, membrane flux, and permeability (Germain et al., 2005; Metzger et al., 2007). TMP is described as the pressure at the inlet P_{in} and at the outlet P_{out} of the inner casing, and at the outlet of the outer casing, P_{perm} (shown in Eq. (2.1)) (Vinther et al., 2014).

$$P_{TMP} = \frac{P_{in} + P_{out}}{2} - P_{perm} \tag{2.1}$$

Membrane flux is the permeate flow per unit area of the membrane and relies on hydraulic resistance, cake layer thickness, and driving force (Izadi et al., 2018). The driving force is the gradient of potential membrane area of mass transport associated with pressure and particle concentration. The mechanism of mass transport in the membrane operation is related to the materials and structure of the membrane (Izadi et al., 2018). Flux also determines the fouling rate or the rate of increase in TMP with time, which indicates how often the membrane should be cleaned and the methods adopted for cleaning (Judd, 2016). For a porous membrane system, Eq. (2.2) is used to estimate the permeate flux, which is proportional to hydraulic resistance.

$$J\left(\frac{L}{m^2 \cdot h}\right) = \frac{TMP}{\mu \cdot R_t} \tag{2.2}$$

where J is permeate flux ($L/m^2 \cdot h$), TMP is transmembrane pressure (kPa), μ is the viscosity of the permeate (Pa.s), R_t is the total resistance (1/m) which is shown in Eq. (2.3). This equation indicates resistance-in-series (RIS) model which is the most comprehensive and applied model in the membrane filtration systems. This model is straightforward and directly associated with the phenomena in the study (Di Bella & Di Trapani, 2019).

$$R_t = R_m + R_c + R_p \tag{2.3}$$

where R_m is the intrinsic membrane resistance, R_c is the cake resistance formed by the cake layer (reversible fouling), and R_p is pore blocking resistance caused by solute adsorption into the membrane pores and gel formation (irreversible fouling) (Psoch & Schiewer, 2006). Permeability is specified as flux per TMP which is another important factor to be considered in membrane filtration systems as calculated by Eq. (2.4) (Izadi et al., 2018).

$$Permeability = \frac{J}{TMP} \tag{2.4}$$

where J is membrane flux and TMP refers to transmembrane pressure.

Hong et al. (2003) realized that increasing TMP declined permeate flux due to increasing the particles transportation to the membrane, and consequently caused particles to be accumulated on the membrane surface. Besides, increase in permeate flow rate intensified drag force and cake layer resistance due to cake layer compression which led to TMP increase.

Membrane technology can operate two ways: constant TMP or constant flux. In a constant-TMP mode, a fast flux decrease is observed at the beginning of the filtration followed by a slow decline until a steady-state flux is obtained while in a constant flux, severe membrane fouling does not occur (Le-Clech et al., 2006). Under a constant flux, fouling in membrane filtration systems happens in a three-stage mechanism. The first stage of membrane fouling has been considered as a conditioning fouling during which there is an initial short-term rise in the TMP. The second stage of membrane fouling is called slow fouling, during this stage the TMP gradually increases linearly or exponentially. The third stage of membrane fouling is associated with a sharp rise in the TMP and also sudden increase in the rate of changes (Zhang et al., 2006).

The majority of previous membrane filtration studies for treatment of oily wastewater used constant TMP (Norouzbahari et al., 2009; Salahi et al., 2010, 2011; Mahbouba et al., 2021), Badrnezhad & Beni (2013) investigated the performance of a UF membrane to treat produced water. In this experiment, optimum operating parameters were temperature of 40˚C , pH of 7.0, and TMP of 3 bar and results showed that oil removal efficiency was over 99%. In this study, the permeate flux decreased rapidly at the beginning of the experiment and it gradually reached a pseudo-steady condition as time passes.

Another effective parameter is crossflow velocity (CFV), which is calculated by dividing the volumetric flow rate through the membrane by the cross section area of the membrane

(Sterlitech Corporation, 2017). Increasing CFV increases turbulence and Reynolds number inside the system and increases shear stress on the membrane surface. Consequently, it reduces the accumulation of substances on the membrane surface, and therefore, improves the performance of the membrane (Masoudnia et al., 2014).

2.9.3. Effect of Aeration

Aeration provides a homogenous distribution of influent inside the membrane tank and decreases the deposition of substances on the surface of the membrane (Luis, 2018). Aeration leads to shear stress and fluctuations on the membrane surface, which is an effective fouling management strategy (Etemadi et al., 2020). When membrane is air scoured, the generated air bubbles are less dense than the surrounding mixed liquor and they move to the water surface as a result of buoyancy forces. The mixed liquor is drawn into the bubble wakes as the bubbles moving upwards and also displaced by rising air bubbles. This gas/liquid two-phase flow has greatly improved the performance of the membrane process compared to increasing liquid flow alone (Drews, 2010). As an example, Wang et al. (2009) used a UF membrane filtration system to treat oily wastewater. In this system, aeration was performed for 2 min at 10-minute filtration intervals. The constant aeration maintained TMP at a low amount and reduced sharp TMP increase.

2.10. Membrane Fouling Mitigation Methods

Cleaning procedures are classified as either in-situ or ex-situ methods depending on where the membrane module is located throughout the cleaning process. The membrane module remains in the membrane tank during the cleaning process in in-situ cleaning, also known as online cleaning. Cleaning the membrane out of the membrane tank (i.e., in another tank outside the membrane filtration unit) is termed as ex-situ procedure or offline cleaning (Zsirai et al., 2012).

2.10.1. Physical Cleaning

Physical cleaning methods rely on mechanical techniques to eliminate foulants from the membrane surface and improve cleaning efficacy. Backwashing, air sparging, relaxation, vibration, and sponge ball cleaning are examples of physical cleaning. Physical methods are effective for removing the cake layer that forms on the membrane surface and eliminating reversible contaminants on the membrane surface or in the membrane pores (Peng & Tremblay, 2008). Physical cleaning is simple because no chemicals are needed, no chemical wastes are produced, and the probability to chemically degrade the membrane is less (Judd, 2010).

Backwashing is the most common method for addressing membrane fouling in which the direction of the permeate flow is reversed to remove foulants deposited on the membrane surface or inside the pores. Combining backwash with air scouring is a common physical method to remove fouling in submerged systems (Hilal et al., 2005). Chua et al. (2003) investigated the influence of backwash duration on the membrane fouling in a pilot-scale HF membrane unit. The results showed that longer backwash duration was more effective than prolonged air scouring in controlling membrane fouling. The process improvement was observed when backwash flow rate increased up to twice that of the permeate flow rate, but further increase in the backwash flow rate did not have any improvements. Ye et al. (2010) investigated the impact of aeration during backwash on the membrane fouling for seawater filtration. That study found that backwash with a moderate air flow rate had a lower final TMP and postponed the fouling rate more efficiently in comparison with a high air flow rate. High air flow rate constrained the air scouring benefits and did not improve reversibility.

Relaxation occurs when the filtration process is stopped while the air scouring of membrane is in progress. This method is commonly conducted in submerged membrane filtration systems for removing reversible fouling. Longer relaxation can be more effective; however, a too long and a highly frequent relaxation would cause severe fouling because of the relatively high instantaneous flux (Wu et al., 2008; Judd, 2010; Yusuf et al., 2016).

Vibration is another method to mitigate membrane fouling in which shear stress is generated on the membrane surface through the relative motion between the membranes and liquid (Li et al., 2014). Ullah et al. (2011) used periodic vibration to dislodge oil droplets from the membrane surface and did not report significant decline in flux which enhanced the membrane performance.

Sponge cleaning is another method to remove membrane fouling; Sadeghi et al. (2017) developed a membrane filtration system to treat saline petrochemical wastewater. Physical cleaning was conducted by rinsing the membrane surface with tap water, and then, wiping it with a soft sponge which was found effective. Takadono et al. (1984) reported that membranes equipped with the automatic sponge ball cleaning systems could successfully treat solutions containing high concentrations of dissolved or suspended solids. This system maintained a high membrane flux under constant TMP improving the efficiency of the membrane filtration unit.

2.10.2. Chemical Cleaning

When membrane fouling becomes severe which is difficult to remove through physical methods, chemical cleaning turns out to be necessary. Chemical cleaning is more effective than physical cleaning in removing foulants attached into the pores of the membrane (Judd, 2010). Chemical cleaning involves immersing a polluted membrane in a chemical cleaning agent that reacts chemically with impurities to remove pollutants from the membrane surface or pores and restore membrane flux (Gul et al., 2021). There are normally six stages to chemical cleaning: (1) bulk dispersion and production of cleaning agents; (2) transport of cleaning agents to the fouled surfaces; (3) transmission through fouled layers; (4) cleaning reactions; (5) transport of reaction products back to the interface; and (6) transport of products back to the bulk solution (Porcelli & Judd, 2010). Some chemicals that are used to remove organic foulant in oily wastewater are bases, oxidants, acids, and surfactants. Each chemical affects the foulant through a different mechanism; for instance, when base is used for chemical cleaning, the associated reactions include hydrolysis, solubilization, saponification, and chelation (complexion with metals) which occur between base and foulants to effectively eliminate the membrane foulants. When acids such as hydrochloric acid (HCl), sulphuric acid (H_2SO_4), nitric acid (HNO_3), and phosphoric acid (H_3PO_4) are used, the reactions are solubilization, hydrolysis, precipitation, dissolution of inorganics, and chelation (Ullah et al., 2021). Therefore, different chemicals can recover membrane flux, decrease TMP, and consequently improve the membrane performance.

Various studies have evaluated chemical cleaning to recover membrane permeability, and reduce membrane fouling, for instance, Huang et al. (2015) used a PVDF-UF membrane filtration system to treat oily wastewater under a constant TMP. In this study, pure water and 3 wt.% sodium hydroxide (NaOH) aqueous solution were used to recover membrane flux. Results showed more than 90% flux recovery which led to membrane fouling mitigation and membrane performance enhancement. In another study, Nyström (1991) used an alkaline solution (NaOH) for cleaning UF membrane when treating oily wastewater; the results showed that chemical cleaning was able to significantly restore the initial water flux.

2.11. Membrane Filtration Application in Oily Wastewater Treatment

Different studies have been conducted to evaluate MF and UF membranes in the treatment of oily wastewater using different operational parameters. For example, Rezvanpour et al. (2009) used flat-sheet cellulose UF membranes to treat oily wastewater containing kerosene. They investigated the effects of TMP, initial oil concentration, feed flow velocity, and pH on the performance of a bench-scale UF system. Results of that study showed that the optimum conditions were TMP of 3 bar and oil concentration of 3% (v/v) which led to the greatest flux (108 $L/m^2 \cdot h$) and 98% of total organic carbon (TOC) removal efficiency. Wang et al. (2009) used a bench-scale membrane unit containing flat-sheet PVDF-MF membranes to treat emulsified oily wastewater. Operational parameters were feed flow rate of 3 ml/min, TMP of 0.2 MPa, and oil concentration of 850 ppm. Results of that study showed that TOC removal efficiency was over 95% and membrane fouling increased with filtration time and decreased the membrane performance. To recover the membrane flux, membranes were chemically cleaned with different chemical solutions. In addition, the fouled membrane was recovered using interval filtration and aeration with the aeration flow rate of 3 ml/min. Findings of the Wang et al. (2009) study showed that the aeration was more effective in maintaining membrane flux and enhancing the membrane performance.

Seyed Shahabadi and Reyhani (2014) studied the performance of a bench-scale flat-sheet PAN-UF membrane filtration system to treat produced water. Different operating parameters such as temperature, TMP, and CFV were considered. The results of that study showed the optimum values for temperature, TMP, and CFV were 50°C, 3.9 bar, and 1.75 m/s, respectively, and oil and grease removal efficiency was 100%. In terms of membrane fouling, high temperature resulted in more oil solubility in water, and therefore, some droplets penetrated the membrane. Oil droplets did not accumulate on the surface of the membrane, and they blocked membrane pores. Increasing CFV enhanced the turbulence of fluid flow which removed the layer of precipitations from the membrane surface. Increasing TMP accumulated more contaminants on the membrane surface which formed a gel layer at higher TMP. This increased flux decline and fouling resistance in the membrane filtration system.

Huang et al. (2015) investigated the performance of a bench-scale flat-sheet PVDF-UF membrane filtration system at TMP of 0.3 MPa for the treatment of oily wastewater. The results of that study showed more than 95% of oil removal efficiency. Fouling mechanism was also examined, and external fouling was the main reason of flux decline. Results indicated that flux recovery of the fouled membranes after chemical cleaning using 3 wt.%

NaOH aqueous solution was more than 90%. Yang et al. (2018) investigated the treatment efficiency of a bench-scale unit containing flat-sheet PES-MF membrane and achieved 98.9% oil removal efficiency under constant TMP.

The selection of the review of the above studies was due to the similarities of their research work to the goal of this thesis to treat oily wastewater; however, they were operated under the constant TMP, and a number of bench-scale studies were more than pilot-scale studies. Most of the published studies reviewed used flat-sheet membranes with a side-stream configuration in physical separation studies to treat oily wastewater when considering the effect of different parameters on the membrane performance (Keyvan Hosseini et al., 2023). Previous UF and MF physical separation studies mostly used TMP, CFV, temperature, pH, and oil concentration as operating parameters and did not provide a thorough analysis about removal efficiencies of different parameters such as various fractions of PHC, and PAHs in oily wastewater (Keyvan Hosseini et al., 2023). The fouling investigation of the majority of previous studies was limited to bench-scale and few of them mentioned the fouling mechanisms and membrane cleaning in the system.

The second group of studies concentrated on HF membrane application for the treatment of oily wastewater. For instance, Zhu et al. (2014) fabricated a modified PVDF-HF membrane to treat oily wastewater containing three types of oil such as hexadecane, crude oil, and palm oil. In an unmodified membrane, the oil contact angle was approximately 15° or lower which indicated the high oleophilicity of the membrane while modified membranes had an oil contact angle of about 75°. Hence, an unmodified membrane adsorbed a higher concentration of oil in comparison with a modified membrane as a result of its hydrophobicity. Results showed that the modified HF membrane had a greater water flux (i.e., 72 $L/m^2 \cdot h$), and less flux reduction (i.e., from 6% to 3%) during oily wastewater treatment. Over 98%, 98%, and 70% oil removal efficiencies for oily wastewater containing hexadecane, crude oil, and palm oil were achieved, respectively. Otitoju et al. (2017) evaluated three different HF membranes under similar operating conditions. The three membranes were PES, PES/SiO_2, and tetraethyloxysilane PES/(TEOS). PES/TEOS indicated the best hydrophilicity compared to the other two membranes. The oil contact angle of the PES/TEOS membrane was 125.47° which was the highest in comparison with PES and PES/SiO_2. This membrane had a 99.98% oil removal efficiency and a permeate flux of 90.937 $L/m^2 \cdot h$, which was higher than two other membranes. This study also revealed that the modified PES/TEOS membrane showed an outstanding antifouling feature and the oil deposition on it was easily washed using physical cleaning. El-badawy et al. (2022) used PVDF-PET braid-reinforced HF membranes to treat oily wastewater. Results showed that flux reached 620 $L/m^2 \cdot h$ and oil removal efficiency was 88% due to high porosity and underwater oleophobicity of the membrane.

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 (Keyvan Hosseini et al., 2022). These studies did not investigate the effect of different operating parameters on the membrane performance. In addition, these experiments were confined to bench-scale laboratory experiments, and the challenge remains in preparing modified HF membranes for commercial use (Hube et al., 2020; Naim et al., 2021).

2.12. Knowledge Gaps and Research Hypothesis

This chapter has laid the basis required for a comprehensive investigation into the performance of a membrane filtration system during the treatment of oily wastewater. Although there have been successful bench-scale and pilot-scale studies on the performance of membranes in treatment of oily wastewater, there have been very few pilot-scale studies applied to the treatment of oily wastewater. Therefore, there is a need to develop a practical experiment which can be used in industrial applications (Yuliwati et al., 2012; Alzahrani & Mohammad, 2014; Hube et al., 2020; Ismail et al., 2020). The majority of the previous studies were operated under the constant TMP which is not appropriate for large-scale applications due to the rapid transportation of particles toward the membrane surface, and consequently severe membrane fouling (Hussain, 2019). The impact of different operating parameters on the membrane performance during the oily wastewater treatment was investigated using flat-sheet membrane rather than HF membrane. HF membranes are more favorable in comparison with flat-sheet membranes due to their higher membrane flux, higher packing density, and lower membrane fouling due to aeration (Akhondi et al., 2017; Altinbas et al., 2021). Membrane filtration studies were mainly performed with side-stream configuration rather than submerged type which is restricted to bench-scale membrane filtration systems (Lenntech, 2022) and tends to severe membrane fouling, and consequently, low membrane performance. Most studies investigated the removal efficiency of TPH, and no studies were found that examine the removal efficiency of different fractions of petroleum and PAHs (Keyvan Hosseini et al., 2023). Among the studies, little information could be found describing the analysis of membrane fouling in a pilot-scale membrane filtration system with heavy and light crude oils. As a result, in this thesis a pilot-scale submerged PTFE-HF membrane filtration unit using UF membranes was used and the analysis of membrane performance in terms of treatment efficiency and membrane fouling was investigated. The effect of operating parameters such as aeration flow rate and fixed membrane flux on hydrocarbon removal efficiency and membrane fouling in the experiments containing heavy and light crude oil was conducted. The following chapter outlines materials and methods, which were used in order to address the research gaps that have been identified.

CHAPTER 3 MATERIALS AND METHODS[1]

This chapter outlines the materials and methods used to assess the performance of a pilot-scale PTFE-HF membrane filtration system to treat synthetic oily wastewater.

First, the characterization of light and heavy crude oils, and the preparation of synthetic oil-in-water emulsion are stated. Second, the methods to analyze different hydrocarbon removal parameters such as TPH, PHC fractions, and PAHs as well as the analysis of fouling resistances are reported followed by statistical analysis. Furthermore, the experimental setup of a pilot-scale membrane filtration system, filtration and cleaning processes, and experimental design approach are discussed.

A research plan was implemented which involved different stages as shown in Figure 3.1. The research plan was split into four phases to obtain the research goals effectively and precisely. These phases consist of preparing oil-in-water emulsion, running a pilot-scale membrane filtration system, performing UF experiments, and conducting analytical and statistical analyses.

---

[1] This chapter is based on the paper that has been published:
Keyvan Hosseini, P., Liu, L., Keyvan Hosseini, M., Bhattacharyya, A., Miao, J., & Wang, F. (2023). Treatment of a Synthetic Decanted Oily Seawater in a Pilot-Scale Hollow Fiber Membrane Filtration Process: Experimental Investigation. Journal of Hazardous Materials, 441, 129928.

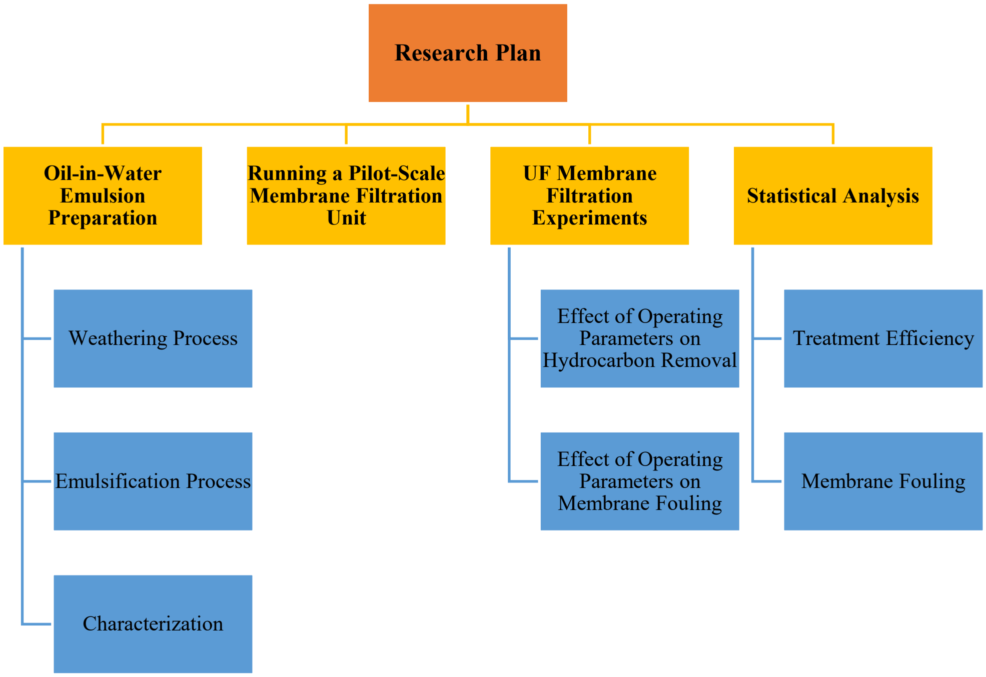

Figure 3.1 Research plan conducted for this thesis

3.1. Preparation of Synthetic Oily Seawater

3.1.1. Crude Oil Characterization

In the experiments, heavy and light crude oil samples such as Cold Lake Dilbit (CLD) and Very Low Sulfur Fuel Oil (VLSFO) were used. The crude oil samples were provided by Multi-Partner Research Initiative (MPRI) in Canada. CLD as a heavy crude oil is a major unconventional product which is transported within North America (Conmy et al., 2017). VLSFO as a light crude oil has recently been approved to be used onboard ships by International Maritime Organization (IMO), which has made it a popular fuel due to its low cost (Kim et al., 2021). These crude oils were selected because both have a high potential to be spilled into the ocean as a result of tanker accidents. The samples from these two types of oil were characterized by the following standard procedures: API gravity

(ASTM D4052), density (ASTM D4052), and kinematic viscosity (ASTM D7042), and the measured quantities are reported in Table 3.1.

Table 3.1 The physical characteristics of two types of crude oil

Oil Type	Density (g/cm^3)	Viscosity (cPs) @ 20℃	API°
CLD	0.924	331	21.8
VLSFO	0.877	10.4	29.7

3.1.2. Oil Weathering Process

The synthetic oily wastewater was prepared to reproduce decanted oily seawater from a marine oil spill clean-up operation in the Atlantic Ocean. In nature, spilled oil is exposed to weathering processes including evaporation, aqueous dissolution, sorption, oxidation, emulsification, and dispersion. Therefore, the chemical composition and physical properties of oil are changed (Yang et al., 2020). The weathering of petroleum products can be replicated in the laboratory by air sparging to remove volatile chemical components (Li et al., 2009). A pressurized air cylinder and tubing were used to simulate the weathering process (shown in Figure 3.2). A pre-weighed container was filled with oil and placed on a balance under the fume hood to measure the weathering rate. The reduction in oil mass indicated the evaporation of volatile components in the oil sample. The rate of evaporation was calculated using Eq. (3.1):

$$R_{evap} = \frac{m_{oil,initial} - m_{oil}(t)}{m_{oil,initial}} \times 100 \tag{3.1}$$

where $m_{oil,initial}$ refers to initial oil weight, and $m_{oil}(t)$ denotes the weight of oil at time t.

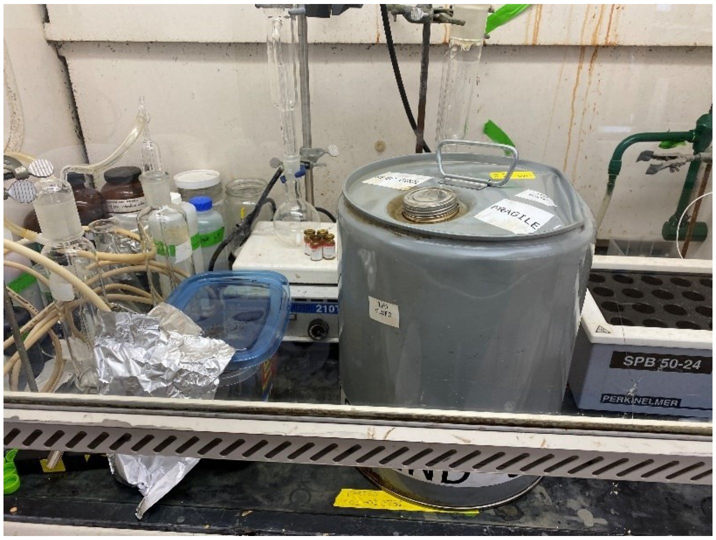

Figure 3.2 Weathering crude oil in the laboratory

3.1.3. Emulsification Process

The oil-in-water emulsion was produced using a high shear vertical mixer (Inoxpa, Spain); during this process the wastewater was drawn from the lower part of the feed tank and the rotor moved it circularly. The oil-in-water emulsion was mechanically and hydraulically sheared at a high speed. The vertical suction and radial thrust both resulted in a flow circulation (Inoxpa, 2021). The schematic diagram of the tank/mixer setup is shown in Figure 3.3.

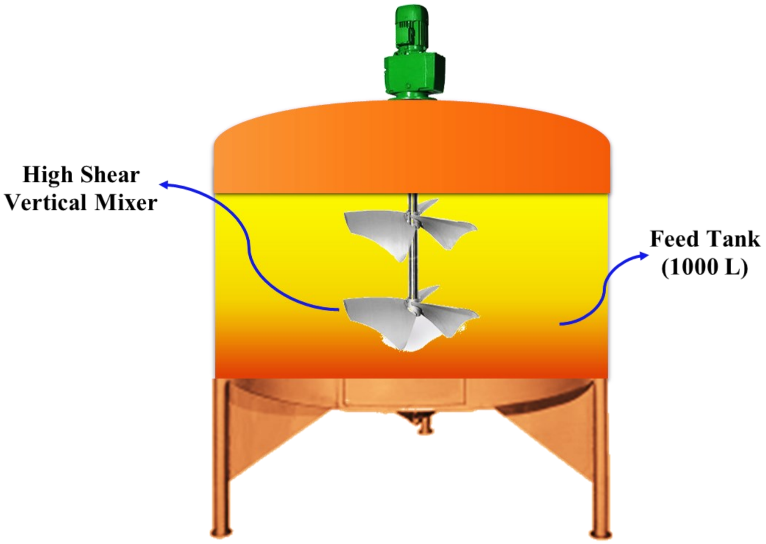

Figure 3.3 The schematic diagram of the tank/mixer set-up for emulsification process

Following weathering process, synthetic oily seawater containing oil-in-water emulsion was prepared using a 1000 L of seawater and mixed with two types of crude oil (i.e., CLD and VLSFO) using different oil concentrations under 3515 rpm. Seawater was provided by Aquatron laboratory (located in Halifax, NS, Canada) which accesses seawater from the Northwest Arm of the Atlantic Ocean. Seawater passed through four sand filters and was exposed to UV treatment to remove suspended solids and bacteria.

3.2. Analytical Methods

3.2.1. Synthetic Oily Seawater Characterization

The determination of the oil droplet size was carried out using a laser light scattering system (Mastersizer, United Kingdom). A laser beam passed through a sample and measured the intensity of light scattered throughout the sample. Deionized water was applied as a dispersive phase.

The electrical conductivity of solutions, total dissolved solids (TDS), and salinity were measured with an Ecosense EC300A handheld which was calibrated for measurements. pH was measured by ROSS Ultra, Thermo Fisher Scientific probe. The first step was to calibrate the pH meter by measuring the pH of standard liquids at acidic pH of 3, neutral pH of 7, and basic pH of 10. After the calibration process, the pH probe was rinsed properly with deionized water. The pH of the sample was measured by immersing the pH probe into the beaker filled with the sample. The determinations of solutions viscosity at room temperature were performed using ASTM D7042.

3.2.2. Treatment Efficiency Analyses

The TPH concentrations of the feed and permeates were measured by standard methods. TPH rejection percentage through the membrane is calculated based on Eq. (3.2) (Radjenovic et al., 2008):

$$R_{rej} = \frac{c_f - c_p}{c_f} \times 100 \tag{3.2}$$

where C_f is TPH concentration in the feed and C_p is its concentration in the permeate after the filtration process.

Ultraviolet-visible (UV-Vis) spectrometry was used to measure TPH; a 10 ml water sample was filled in a centrifuge tube. After that, 1 ml of dichloromethane (DCM) was added. The mixture was vortexed for 60 seconds and settled until the water and DCM were completely separated. The absorbances of 340 nm, 370 nm, and 400 nm were measured by UV-Vis spectrometer and integrated to calculate the TPH concentration of the organic phase according to methodology outlined in Zheng et al. (2015) and Song et al. (2022).

PHCs are comprised of readily volatile decanes (F1: C6-C10), semi-volatile hexadecanes (F2: C10-C16), non-volatile tetratriacontane (F3: C16-C34), and non-volatile pentacontane (F4: C34-C50). These fractions have different toxicity mechanisms; F1 and F2 as the lighter fractions have relatively high-water solubility and bioactivity which lead to acute toxicity. F3 and F4 are a complex combination of aromatic, aliphatic, heterocyclic, and asphaltene hydrocarbons that are very hydrophobic and recalcitrant which make them unable to break down (Wang, 2008). PAHs are a subdivision of PHCs which are persistent organic compounds. These components are mutagenic and carcinogenic, and do not degrade easily under natural conditions. The USEPA has recorded 16 PAHs as priority contaminants (Rutter et al., 2014).

PHCs were evaluated by gas chromatography/mass spectrometry (GC-MS); a 10 ml water sample was filled in a centrifuge tube. Following that, 1 ml of DCM was added, and then, the mixture was vortexed for 60 seconds and settled until the water and DCM were fully separated. 100 µL of the organic phase was transferred to a 150-µL vial and then 10 µL of internal standard was added. The mixture was analyzed through an Agilent 7890A gas chromatogram-mass spectrum system. A 30 m DB-5ms capillary GC column was used, and the carrier gas was Helium. The GC oven temperature was set at 45 °C for 2 min, then ramped up 6 °C/min to 300 °C for 20 minutes. A selected ion mode was used to obtain a better signal to noise (S/N). The GC-detectable n-alkanes C8-C38 alkanes (F1-F4) and PAHs were profiled and identified through specific mass/charge number (m/z) values using the ChemStation Software (Zheng et al., 2015).

3.2.3. Membrane Fouling Analysis

The fouling resistances were calculated using a four-step procedure as outlined by Di Bella and Di Trapani (2019): (1) R_m was measured by calculating the TMP in clean water; (2) R_t was evaluated on the basis of the final TMP value before the last backwash; (3) R_m+R_p was evaluated based on the final TMP after the last backwash. After a backwash was conducted, the membrane was subjected to filtration cycle for 9 minutes and R_p was derived by subtracting R_m obtained from step 1. 4) R_c was determined from the R_t achieved in step 2. TMP was recorded every minute on the pilot-scale membrane system using a PLC control panel (WE!NVIEW, China).

3.3. Statistical Analysis

Statistical analyses were performed to evaluate major factors affecting removal efficiency and membrane fouling in the membrane filtration system. The statistical analysis was carried out using MINITAB 19 where analysis of variance (ANOVA) test was conducted.

3.4. Experimental Setup and Design

The schematic design drawing of the membrane filtration system is shown in Figure 3.4. The entire pilot-scale system has dimensions of 2280 mm × 914 mm × 2000 mm (L × W × H).

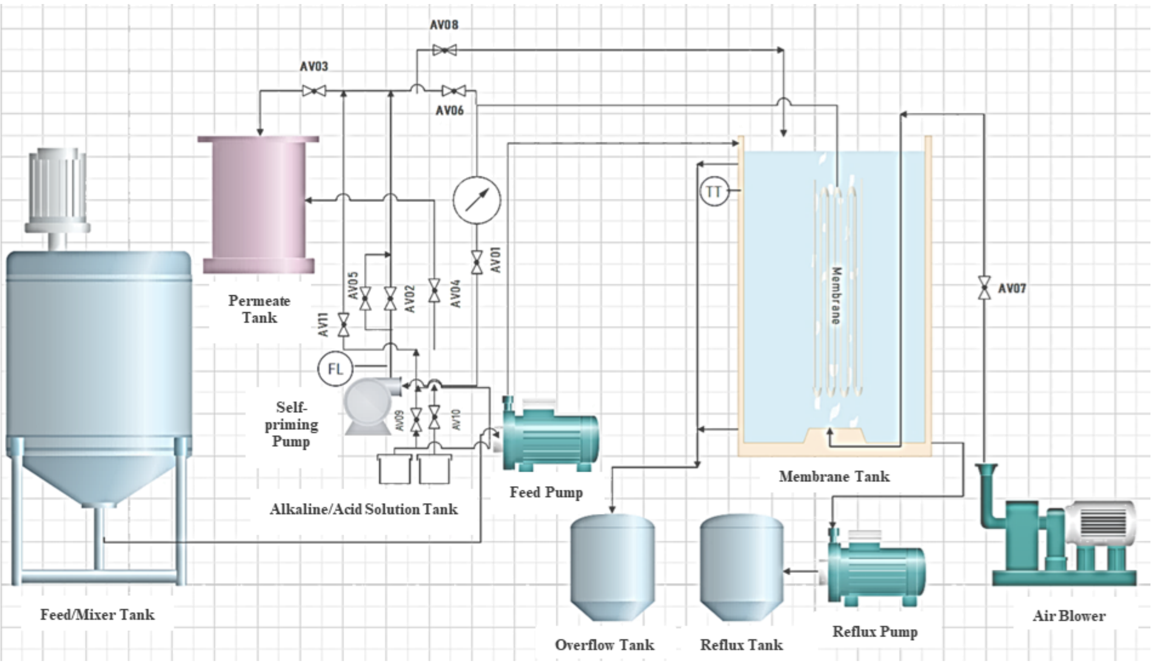

Figure 3.4 Schematic design drawing of the pilot-scale membrane filtration unit

The membrane tank has a working volume of 340 L and two HF-PTFE membrane modules with a nominal pore size of 0.08 µm (Sumitomo Electric, Japan). PTFE is used extensively in different fields of industries due to high thermal stability, superior chemical resistance, and high mechanical strength (Xu et al., 2012). The PTFE membranes were exposed to a surface modification by the manufacturer using a hydrophilic polymer which decreases its water contact angle to 58° from 146°. Table 3.2 shows the key specification of the PTFE membrane modules used in this study.

Table 3.2 The specification of PTFE membrane modules (Sumitomo Electric)

Parameter	Value (unit)
Material	PTFE
Nominal Pore Size	0.08 µm
TMP (Filtration)	> 40 kPa

Maximum Operating Temperature	50 °C
TMP (Backwash)	< 100 kPa
Length	1300 mm
Bottom Section	154 × 164 mm
Membrane Area	6 m^2

3.4.1. Filtration and Cleaning Processes

Synthetic oily seawater was pumped from the feed tank to the membrane tank using a feed pump (Dongguan Riyiguobao, MPX-250-C-C-V-5-V38-A). The pilot-scale unit was operated continuously at specific inlet flow rate (i.e., UF experiments: inlet flow rate of 1500 L/h). The filtration process consisted of a cycle of 9 minutes of filtration and 1 minute of relaxation and after three cycles a 15-second backwash happened based on the membrane manufacturer's suggestion. The filtration and cleaning procedures are shown in Figure 3.5. The filtration and backwash processes were conducted with a self-priming pump (Dongguan Riyiguobao, MPX-250-C-C-V-5-V38-A).

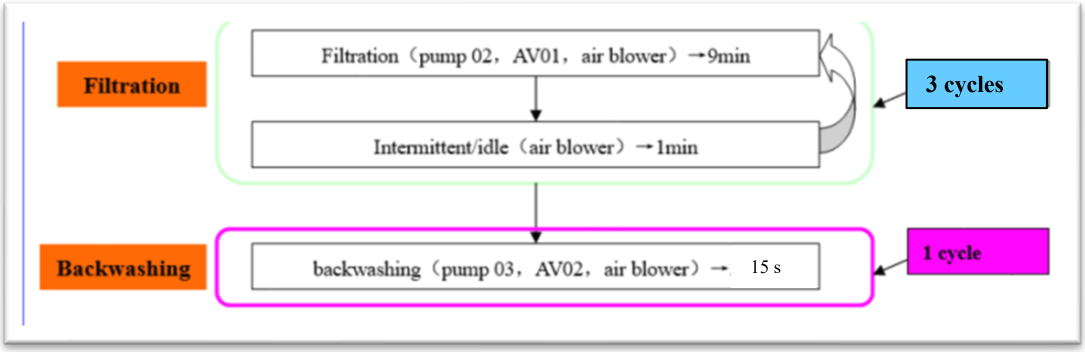

Figure 3.5 Filtration and cleaning procedures

This pump was capable of changing flow direction as required using automatic electromagnetic valves in the system. Aeration was performed using an air blower (HCC-301S) at the bottom of the membranes which was constant throughout the experiments to reduce membrane fouling and promote the recirculation of liquid within the tank. The air flow rate was adjusted with manual valves and monitored through an air rotameter which was placed inside the system. To avoid overflowing of the membrane tank, a pipe at the bottom of the membrane tank led to a reflux pump (Dongguan Riyiguobao, MPX-250-C-C-V-5-V38-A) to return water to the feed tank based on the inlet flow rate. TMP from the pressure gauge and filtration flow rate from the electromagnetic flow meter were recorded automatically every minute with the help of a PLC during each run.

This system was equipped with online chemical cleaning and the instructions were provided by the membrane manufacturer. Chemical cleaning was conducted before changing the oil type or if TMP reached 40 kPa. The membrane filtration unit had two chemical cleaning tanks with working volumes of 60 L each, one for acid and one for alkaline solution. The membrane manufacturer recommended mixing 300 to 3000 mg/L sodium hypochlorite (NaClO) and 100 to 500 mg/L NaOH for removing organic foulants and 300 to 3000 mg/L HCl for removing inorganic foulants. In the system, 300 mg/L and 100 mg/L NaClO and NaOH were used, respectively followed by 300 mg/L HCl. After adding the chemical through the membrane into the membrane tank, it was left to soak for 2 hours for each, alkaline and acid cleaning. The membrane unit was then flushed with water to eliminate the chemicals from the system. Waste residuals were drained into a wastewater storage tank before starting the experiments.

3.4.2. Experimental Design Approach

The experiments were designed according to Taguchi method to be able to use its S/N ratio analytical tool. The study was conducted using UF membranes with two levels for membrane flux and aeration flow rate to treat oily seawater containing two types of oil (heavy crude oil (i.e., CLD) and light crude oil (i.e., VLSFO)) using different initial oil concentrations. A design of two factors with two levels resulted in an orthogonal array of L_4 (2^2) and in total 16 experiments were used to assess the effects of operating parameters on the UF membrane performance (shown in Table 3.3).

Table 3.3 Experimental design for UF experiments

<table>
<tr><th colspan="4">CLD Experiments</th></tr>
<tr><th>Experimental Run</th><th>Membrane Flux (L/m^2·h)</th><th>Aeration Flow Rate (m^3/h)</th><th>Oil Concentration (ppm)</th></tr>
<tr><td>1</td><td>6</td><td>4.8</td><td>50</td></tr>
<tr><td>2</td><td>6</td><td>6</td><td>50</td></tr>
<tr><td>3</td><td>12</td><td>4.8</td><td>50</td></tr>
<tr><td>4</td><td>12</td><td>6</td><td>50</td></tr>
<tr><td>5</td><td>6</td><td>4.8</td><td>100</td></tr>
<tr><td>6</td><td>6</td><td>6</td><td>100</td></tr>
<tr><td>7</td><td>12</td><td>4.8</td><td>100</td></tr>
<tr><td>8</td><td>12</td><td>6</td><td>100</td></tr>
<tr><th colspan="4">VLSFO Experiments</th></tr>
<tr><th>Experimental Run</th><th>Membrane Flux (L/m^2·h)</th><th>Aeration Flow Rate (m^3/h)</th><th>Oil Concentration (ppm)</th></tr>
<tr><td>1</td><td>6</td><td>4</td><td>50</td></tr>
<tr><td>2</td><td>12</td><td>4</td><td>50</td></tr>
<tr><td>3</td><td>6</td><td>4.8</td><td>50</td></tr>
<tr><td>4</td><td>12</td><td>4.8</td><td>50</td></tr>
<tr><td>5</td><td>6</td><td>4</td><td>100</td></tr>
<tr><td>6</td><td>6</td><td>4.8</td><td>100</td></tr>
<tr><td>7</td><td>12</td><td>4.8</td><td>100</td></tr>
<tr><td>8</td><td>12</td><td>4</td><td>100</td></tr>
</table>

The Taguchi ‘signal’ and ‘noise’ indicate the desirable and undesirable value for output characteristics, respectively, and their ratio (S/N) is used to transform responses to optimize the membrane filtration process (Kim et al., 2007). The equation of the S/N ratio depends on the criterion for the quality characteristics to be optimized. Since a high removal efficiency is desired, the “the larger the better” criterion was selected for the S/N ratio. Eqs. (3.3) and (3.4) are used to calculate the S/N ratio and mean square deviation (MSD), respectively (Hesampour et al., 2008; Reyhani et al., 2013).

$$\frac{S}{N} = -10 \log_{10} \frac{1}{MSD} \tag{3.3}$$

$$MSD = \frac{Y_1^2 + Y_2^2 + \ldots + Y_N^2}{N} \tag{3.4}$$

where MSD is the mean square deviation, Y is the response factor, and N is the number of observations.

CHAPTER 4 RESULTS AND DISCUSSION[2]

This chapter outlines the results and discussion related to different stages of this thesis. The results associated with oil weathering and synthetic oily seawater characteristics are first reported to provide a better understanding about the synthetic oily seawater. Next, the results of UF experiments pertaining to treatment efficiency and membrane fouling analyses are discussed.

4.1. Characteristics of Synthetic Oily Seawater

The results of the weathering experiments showed that the heavy crude oil (CLD) had a 15% mass loss within 24 h while the light crude oil (VLSFO) underwent a mass loss of 1.5% within 48 h. The light crude oil had significantly lower weathering rate compared to the heavy crude oil. This would be due to the fact that VLSFO was sufficiently weathered before receiving and most of its volatile components were previously weathered.

The oil-in-water emulsion droplet size analysis showed that most of VLSFO oil droplets in the emulsified samples were in the range of 1 to 10 μm in diameter through high shear vertical mixer. This range of oil droplets indicates stable condition of oil-in-water emulsion (Chakrabarty et al., 2008). The results also showed that 90% of the samples containing 50 and 100 ppm oil in seawater had a size of 4.02 (shown in Figure 4.1 (a)) and 7.87 μm (shown in Figure 4.1 (b)) or smaller, respectively. The stable emulsion inside the feed tank

---

[2] This chapter is based on the paper that has been published:
Keyvan Hosseini, P., Liu, L., Keyvan Hosseini, M., Bhattacharyya, A., Miao, J., & Wang, F. (2023). Treatment of a Synthetic Decanted Oily Seawater in a Pilot-Scale Hollow Fiber Membrane Filtration Process: Experimental Investigation. Journal of Hazardous Materials, 441, 129928.

is presented in Figure 4.2. Characteristics of oil-in-water emulsion were tested for feed samples which are shown in Table 4.1.

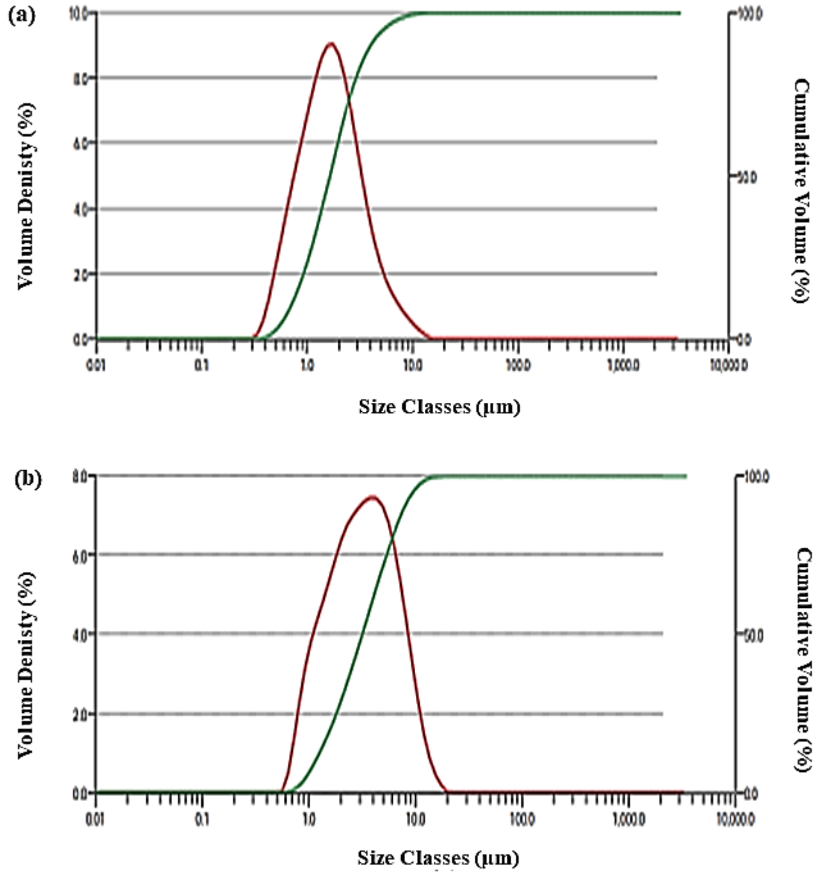

Figure 4.1 Droplet size distribution of oil-in-water emulsion with (a) 50 ppm, (b) 100 ppm

Figure 4.2 Stable oil-in-water emulsion

Table 4.1 Oily wastewater characterization

Conductivity (mS/cm)		39.5
Total dissolved solids (TDS) (g/L)		30.5
Salinity (ppt)		30.5
Oil Concentration (ppm)		CLD: 50, 100 VLSFO: 50, 100
pH		8.2
Viscosity (23℃) (Pa·s)	**50 ppm (CLD)**	0.0009989
	100 ppm (CLD)	0.001044
	50 ppm (VLSFO)	0.0009943
	100 ppm (VLSFO)	0.0009970

4.2. UF Experimental Results

4.2.1. Effect of Aeration on TPH Concentrations of CLD and VLSFO Influents

The initial oil concentrations added to the feed tank were 50 and 100 mg/L; however, the amount of TPH concentrations measured after taking samples from the membrane tank were lower than initial amounts as observed in Table 4.2. These results suggest that air bubbles adhered to the oil and pushed it to the surface, demulsifying the solution which led to rapid reduction of TPH concentration in influent collected from the membrane tank. Similar results were found by Aliff Radzuan et al. (2016) which showed the role of aeration in reduction of TPH concentration in DAF technology. In the system, aeration helped enhance overall removal efficiency of synthetic oily seawater as it operated similar to DAF systems. After each run, the wastewater from the membrane tank along with the permeate was returned to the feed tank for the next run. After discharging the wastewater from the membrane tank, it still maintained about 15 cm of wastewater, in which the majority of the demulsified oil remained. The reflux pump pumped water from the membrane tank back to the feed tank throughout each experimental run and the accumulation of the demulsified floating oil at the top of the membrane tank was observed which was not recirculated back into the feed tank for emulsification. In addition, initial wastewater samples from the membrane tank were collected after 5 min of aeration.

Table 4.2 TPH removal efficiency for oily seawater containing CLD and VLSFO

Experimental Run (CLD)	Membrane Flux (L/m^2·h)	Aeration Flow Rate (m^3/h)	Initial Oil Concentration in the Feed Tank (ppm)	Initial TPH Concentration in the Membrane Tank (ppm)	TPH Concentration in the Permeate Tank (ppm)	TPH Removal (%)	S/N Ratio
1	6	4.8	50	11.95	0.13	98.91	39.90
2	6	6	50	5.62	0.31	94.48	39.51
3	12	4.8	50	3.51	0.11	96.87	39.72
4	12	6	50	2.36	0.22	90.68	39.15
5	6	4.8	100	8.78	0.17	98.06	39.83
6	6	6	100	9.55	0.17	98.22	39.84
7	12	4.8	100	9.99	0.17	98.30	39.85
8	12	6	100	11.12	0.98	91.19	39.2

Experimental Run (VLSFO)	Membrane Flux (L/m^2·h)	Aeration Flow Rate (m^3/h)	Initial Oil Concentration in the Feed Tank (ppm)	Initial TPH Concentration in the Membrane Tank (ppm)	TPH Concentration in the Permeate Tank (ppm)	TPH Removal (%)	S/N Ratio
1	6	4	50	13.3	0.3	97.75	39.80
2	12	4	50	24.35	0.18	99.26	39.94
3	6	4.8	50	17.47	0.18	98.97	39.91
4	12	4.8	50	22.55	0.24	98.94	39.91
5	6	4	100	64.67	0.22	99.66	39.97
6	6	4.8	100	19.09	0.17	99.11	39.92
7	12	4.8	100	27.91	0.2	99.28	39.94
8	12	4	100	18.36	0.24	98.69	39.89

4.2.2. Effect of Membrane Flux and Aeration Flow Rate on TPH Removal Efficiency

The efficiency of TPH removal and the computed S/N ratio for each experimental run are shown in Table 4.2. The S/N ratio was used to specify the optimal operating conditions of the membrane filtration system to obtain the best TPH removal efficiency. It was observed that membrane flux of 6 $L/m^{2}\cdot h$ and aeration flow rate of 4.8 m^3/h resulted in a higher S/N ratio for the experiments performed with CLD as shown in Figure 4.3 (a, b). Membrane flux of 12 $L/m^{2}\cdot h$ and aeration flow rate of 4.8 m^3/h for 50 ppm of VLSFO resulted in a higher S/N ratio, whereas a membrane flux of 6 $L/m^{2}\cdot h$ and aeration flow rate of 4.8 m^3/h in 100 ppm of VLSFO resulted in a higher TPH removal efficiency as shown in Figure 4.3 (c, d). The main effect plot for S/N ratios of TPH removal efficiency is shown in Figure 4.3.

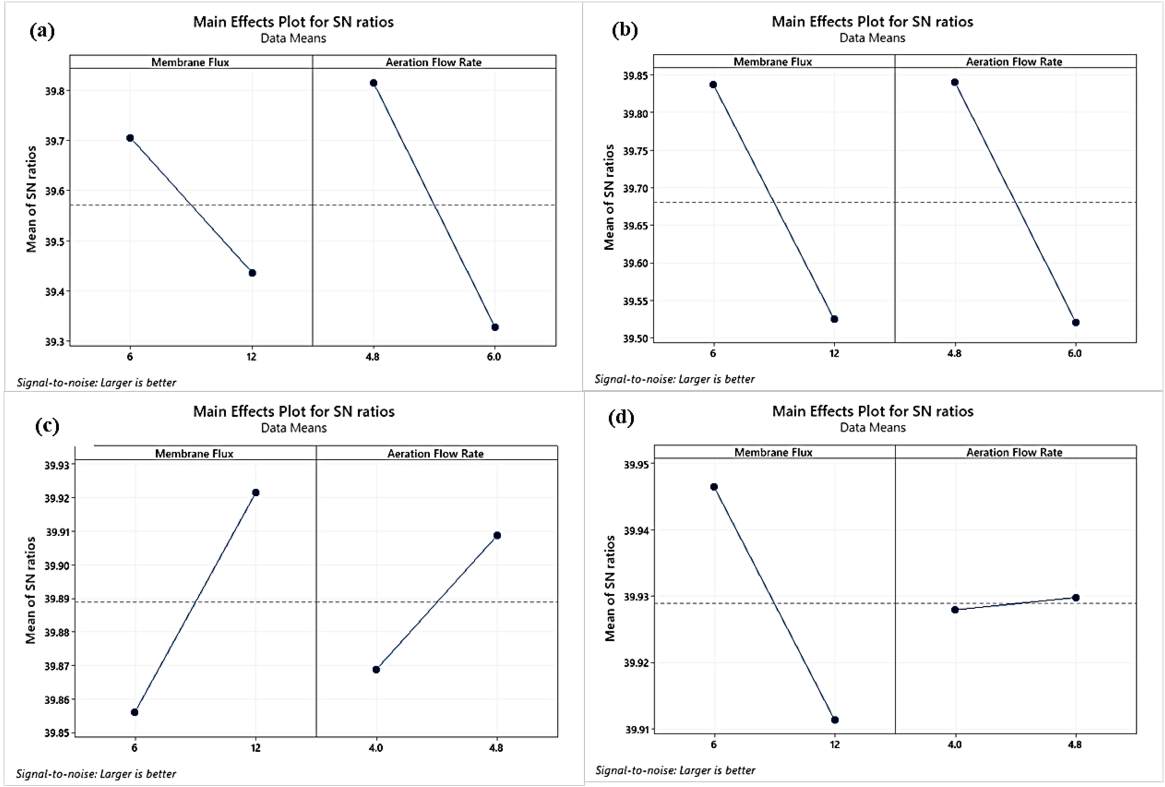

Figure 4.3 Main effect plot for S/N ratios of TPH removal efficiency for two factors at two levels with (a) 50 ppm CLD, (b) 100 ppm CLD, (c) 50 ppm VLSFO, (d) 100 ppm VLSFO

The absence of oil recirculation in the membrane tank led to the accumulation of demulsified oil at the surface of the membrane tank as a result of the aeration in the system; consequently, aeration functioned as a DAF in this system which improved oil removal efficiency. Runs 2, 4, 6, and 8 with aeration flow rate of 6 m^3/h compared to runs 1, 3, 5, and 7 with aeration flow rate of 4.8 m^3/h in CLD experiments indicated that increasing aeration flow rate caused formation of larger air bubbles and enhanced the flotation process inside the system. A large quantity of bubbles increases the possibility of collisions and adhesions among bubbles and oil droplets. Increasing aeration flow rate increases the bubble size. Bubble coalescence process promotes large bubbles formation in the flotation tank. As the bubble size increases, the number of bubbles decrease, which leads to a decrease in oil removal efficiency in the system (Huang and Long, 2020). This was observed in CLD experiments while the difference in oil removal efficiency between 4.8 m^3/h and 4 m^3/h in VLSFO was not reflected because of the small difference in aeration flow rates.

In the experiments, lower membrane flux led to higher removal efficiency compared to higher membrane flux. This is because low membrane flux gives more time for oil-in-water emulsion to be filtered by UF membranes. Convection allows oil to be dragged across a membrane due to the existence of a pressure gradient (Huang et al., 2012). Low membrane flux represents low convection transport leading to low surface tension forces between water and oil. Therefore, it leads to the low mass transfer, and consequently low movement of oil across the membrane and reduces the passage of oily wastewater through the membrane. Thus, lower membrane flux enhances the quality of the permeate during filtration (McCabe et al., 1993). Among different runs of CLD, run 5 (i.e., aeration flow rate of 4.8 m^3/h and membrane flux of 6 $L/m^2 \cdot h$) had slightly lower TPH removal efficiency in comparison with run 7 (i.e., aeration flow rate of 4.8 m^3/h and membrane flux of 12 $L/m^2 \cdot h$) which was conducted at a higher membrane flux. One possible reason for the lower TPH removal efficiency could be due to oil sticking to the sides of the tank. The analogous results were achieved from VLSFO runs and confirmed that lower membrane flux was more effective in TPH removal efficiency. Runs 3 (i.e., aeration flow rate of 4.8 m^3/h and membrane flux of 6 $L/m^2 \cdot h$) and 5 (i.e., aeration flow rate of 4 m^3/h and membrane flux of 6 $L/m^2 \cdot h$) with lower membrane flux showed higher TPH removal efficiency; however, runs 1 (i.e., aeration flow rate of 4 m^3/h and membrane flux of 6 $L/m^2 \cdot h$) and 6 (i.e., aeration flow rate of 4.8 m^3/h and membrane flux of 6 $L/m^2\cdot h$) showed negligibly lower TPH removal in comparison with the corresponding runs with higher membrane flux which would be due to sticking oil to the walls of the tank.

ANOVA analysis was conducted for TPH removal efficiency of CLD and VLSFO and the results were given in Table 4.3.

Table 4.3 ANOVA results for TPH removal efficiency with oily seawater containing CLD and VLSFO

TPH Removal Efficiency of CLD

Source	Degree of Freedom	F-Value	P-Value
Regression	3	5.19	0.073
Oil concentration	1	0.74	0.438
Membrane flux	1	5.05	0.088
Aeration flow rate	1	9.78	0.035
Error	4	-	-
Total	7	-	-

TPH Removal Efficiency of VLSFO

Source	Degree of Freedom	F-Value	P-Value
Regression	3	0.47	0.721
Oil concentration	1	1	0.374
Membrane flux	1	0.14	0.728
Aeration flow rate	1	0.27	0.633
Error	4	-	-
Total	7	-	-

CLD results show that p-value of aeration flow rate was less than 0.05 indicating the importance of this parameter on TPH removal. However, none of the parameters had a significant impact on the TPH removal efficiency in VLSFO experiments ($p\text{-value}>0.05$). This was due to the fact that initial oil concentrations in the membrane tank were much lower than the initial oil concentrations in the feed tank and the selected aeration flow rates were relatively close to each other, which led to very similar oil removal efficiencies for all the runs. The F-value for CLD was 5.19 indicating the model was significant. There was 7.3% chance that a “Regression Model F-Value” this large could occur due to noise. While for VLSFO, F-value was 0.47 which was not significant due to 72.1% of noise.

The closer the R^2 value is to 1, the more precise the relationship between the independent variables and the response. As the R^2 of the model in CLD runs was equal to 0.7956, only 20.44% of the total variance could not be explained by the proposed regression model (Eq. (4.1)).

VLSFO regression model (Eq. (4.2)) was not precise since the p-value was higher than 0.05 and R^2 of the model was equal to 0.2597. This could be described by the low variation in oil removal efficiencies between the different runs and lack of replication in the experiment.

$$TPH_{Removal\ Efficiency,\ CLD} = 118.53 + 0.0241C_{oil} - 0.526Q_{membrane} - 3.66Q_{air} \tag{4.1}$$

$$TPH_{Removal\ Efficiency,\ VLSFO} = 96.73 + 0.00910C_{oil} + 0.0283Q_{membrane} + 0.294Q_{air} \tag{4.2}$$

where C_{oil} refers to oil concentration (ppm), $Q_{membrane}$ is the membrane flux (L/m^2·h), and Q_{air} is the aeration flow rate (m^3/h).

4.2.3. PHCs Removal in CLD and VLSFO Experiments

Figure 4.4 shows the concentrations of PHCs in influent and permeate of different runs for CLD. Most of the F1 fractions from two types of oil were removed during the oil weathering process as a result of its volatile nature. In some CLD runs such as 1, 2, 3, 4, 6, and 8 with membrane fluxes of 6 and 12 $L/m^2 \cdot h$ and aeration flow rates of 4.8 and 6 m^3/h, the concentration of F1 fraction increased in permeate compared to influent. It was observed that the effect of membrane flux and aeration flow rate in different runs on F1 fraction removal efficiency was not recognized. This would be as a consequence of sampling errors and/or volatility of this fraction. In CLD runs, the removal efficiency of F3 was the highest compared to the other fractions, different runs indicated that aeration flow rate of 4.8 m^3/h and membrane flux of 12 $L/m^2 \cdot h$ resulted in higher F3 removal efficiency. F2 removal efficiency was higher in the runs with aeration flow rate of 12 m^3/h and membrane flux of 6 $L/m^2 \cdot h$. Using aeration flow rate of 6 m^3/h and membrane flux of 6 $L/m^2 \cdot h$ led to higher F4 removal efficiency. The results showed that higher membrane flux and higher aeration flow rate were more effective in removing PHC fractions in CLD runs. This would be as a result of the complex nature of each petroleum hydrocarbon fractions or sampling errors. The results also showed that the removal efficiency of F3 fraction in all runs was the highest, followed by F2, F1, and F4, respectively (shown in Figure 4.4).

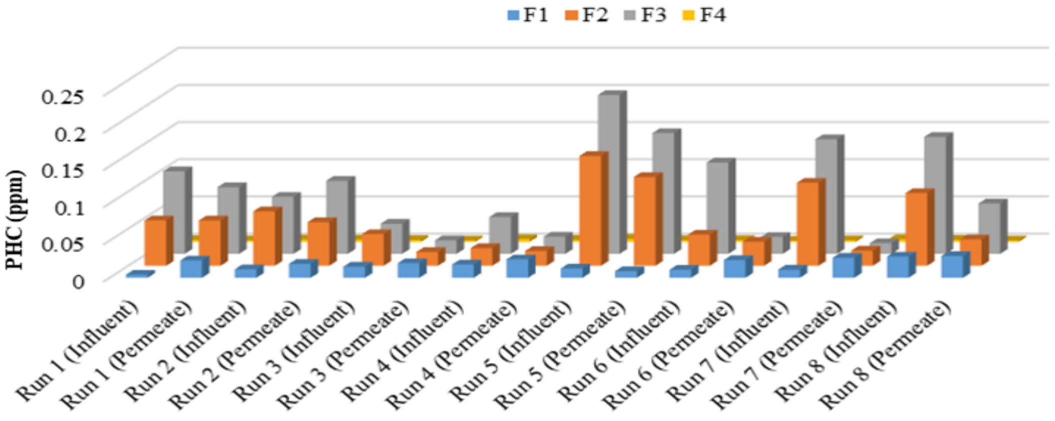

Figure 4.4 Influent and permeate concentrations of PHC in CLD experiments

Figure 4.5 shows the concentrations of PHCs in influent and permeate of different runs for VLSFO. In VLSFO experiments, runs with aeration flow rate of 4 m^3/h and membrane flux of 6 L/m^2·h resulted in higher removal efficiency of both F2 and F1. F3 had higher removal efficiency in the runs with aeration flow rate of 4.8 m^3/h and membrane flux of 6 L/m^2·h. Comparing different VLSFO runs showed that using aeration flow rate of 4 m^3/h and membrane flux of 6 L/m^2·h resulted in higher removal of petroleum hydrocarbon fractions. The removal efficiency of F2 fraction was the highest which can be associated with its volatile nature throughout the filtration process (shown in Figure 4.5). F3 also had an equally high removal efficiency. Comparing the results of two types of oil showed that the effect of operating parameters on the removal of PHC fractions was different depending on the condition of the experiments (i.e., oil physical features, volatility of components, toxicity mechanism, sampling errors, and sticking oil to the walls of the tanks).

The high removal efficiency of F3 would be attributed to the poor solubility of this fraction in water (ChemSafetyPro, 2021). For example, the aromatic components have a water solubility of between 6.6-650 µg/L (Polak & Lu, 1973; Maxxam, 2021), and aliphatic components have a water solubility of 0.0025 µg/L in F3 (Maxxam, 2021). Since the hydrocarbons in F3 fractions are extremely hydrophobic, they are repelled by the hydrophilic Sumitomo PTFE membranes (water contact angle of 58˚). Consequently, most F3 fractions in runs with CLD and VLSFO are retained in the membrane tank, thus a high removal efficiency was achieved.

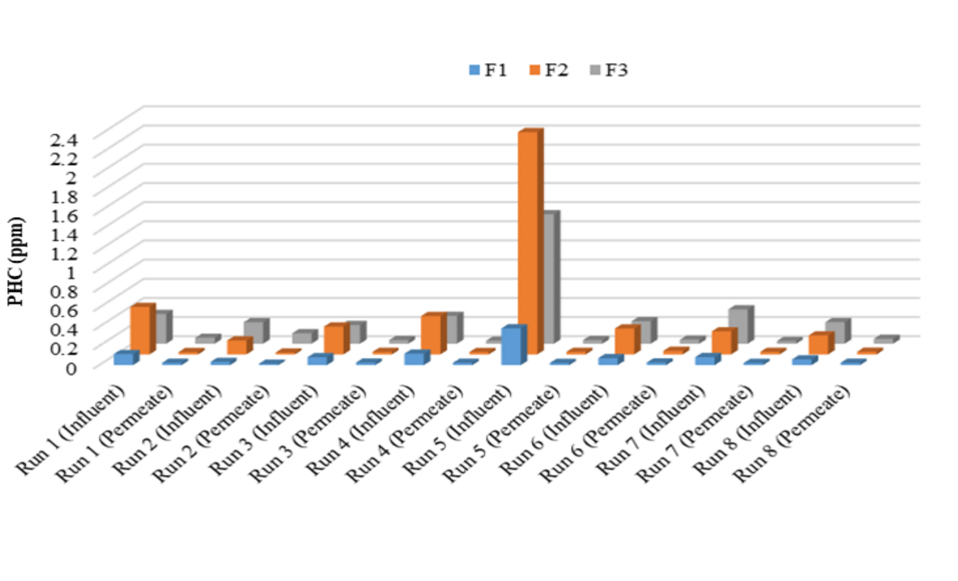

Figure 4.5 Influent and permeate concentrations of PHC in VLSFO experiments

4.2.4. PAHs Removal in CLD and VLSFO Experiments

Different PAH compounds were investigated for CLD experiments, and the results showed that all PAH concentrations were less than 1 ppb. The results of PAHs for runs using VLSFO are shown in Table 4.4. 18 PAHs such as naphthalene, 1-methylnaphthalene, 2-

methylnaphthalene, acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, pyrene, fluoranthene, benzo (a) anthracene, chrysene, benzo (b) fluoranthene, benzo (e) pyrene, benzo (a) pyrene, indeno (1, 2, 3-cd) pyrene, dibenzo (a, h) anthracene, and benzo (g, h, i) perylene were investigated in this study.

The solubilities of 1-methylnaphthalene, 2-methylnaphthalene, anthracene, fluorene, phenanthrene, fluoranthene, acenaphthene, chrysene, and pyrene are 25000, 24600, 4340, 1680-1980, 1200, 200-260, 3930, 2000, and 135 µg/L, respectively (Inchem, 2006; Government of Canada, n.d.a, n.d.b; Syafiuddin & Boopathy, 2021; National Center for Biotechnology Information, 2022a, 2022b, 2022c, 2022d, 2022e, 2022f). All quantities are less than 100000 µg/L which makes them relatively insoluble. Hydrophobicity is a result of low solubility which causes the PAH compounds to be repelled by the hydrophilic membrane, and this leads to a high removal efficiency. The values of PAH in acenaphthylene, benzo (b) fluoranthene, benzo (e) pyrene, benzo (a) pyrene, benzo (a) anthracene, indeno (1, 2, 3-cd) pyrene, dibenzo (a, h) anthracene, and benzo (g, h, i) perylene were negligibly detected in the influent and permeate (Syafiuddin & Boopathy, 2021). The membrane filtration system containing PTFE-UF-HF membranes effectively removed PAH compounds reducing the oil toxicity.

Table 4.4 Concentrations of PAHs in oily seawater containing VLSFO

	Run	1	2	3	4	5	6	7	8
Compound		**PAHs concentration (ppm)**							
Naphthalene	Influent	1.643	0.578	0.951	0.881	4.155	0.943	0.833	0.829
Naphthalene	Permeate	0.363	0.258	0.277	0.256	0.587	0.429	0.318	0.356
1-MethylNAP	Influent	3.742	1.195	1.825	2.161	10.359	2.031	1.833	1.932
1-MethylNAP	Permeate	0.717	0.483	0.472	0.430	1.146	0.542	0.646	0.671
2-MethylNAP	Influent	2.336	0.832	1.177	1.367	6.474	1.297	1.221	1.239
2-MethylNAP	Permeate	0.567	0.404	0.391	0.412	0.901	0.469	0.632	0.528
Acenaphthene	Influent	0.249	0.093	0.129	0.155	0.829	0.121	0.134	0.119
Acenaphthene	Permeate	0.068	0.058	0.071	0.151	0.113	0.023	0.051	0.024
Fluorene	Influent	0.493	0.268	0.385	0.437	0	0.319	0.329	0.269
Fluorene	Permeate	0.291	0.159	0.119	0.121	0.223	0.087	0.147	0.087
Anthracene	Influent	3.458	1.905	2.559	3.050	15.674	2.699	3.111	2.257
Anthracene	Permeate	0.564	0	0.775	0.785	1.003	0.533	1.203	0.718
Phenanthrene	Influent	0.341	0	0.242	0.291	1.778	0.249	0.325	0.209
Phenanthrene	Permeate	0	0	0.070	0.046	0.081	0.032	0.173	0.055
Benzo(b)fluoranthene	Influent	0.095	0.030	0	0	0	0	0.045	0
Benzo(b)fluoranthene	Permeate	0	0	0	0	0	0	0	0
Benzo(e)pyrene	Influent	0	0	0	0	0	0	0.034	0
Benzo(e)pyrene	Permeate	0	0	0	0	0	0	0	0

Benzo(a)pyrene	Influent	0	0.014	0	0	0.082	0	0.013	0
Benzo(a)pyrene	Permeate	0	0	0	0	0	0	0	0
Pyrene	Influent	0.256	0.157	0.198	0.258	2.990	0.235	0.311	0.211
Pyrene	Permeate	0	0	0.030	0.029	0.106	0	0.044	0
Fluoranthene	Influent	1.473	0.845	1.117	1.489	8.235	1.482	1.773	1.302
Fluoranthene	Permeate	0	0	0.069	0.077	0	0.078	0.166	0.126
Benzo(a)anthracene	Influent	0.061	0.039	0	0.075	0.325	0.037	0.073	0.047
Benzo(a)anthracene	Permeate	0	0	0	0	0	0	0	0
Indeno(1,2,3-cd)pyrene	Influent	0	0	0	0	0	0	0	0
Indeno(1,2,3-cd)pyrene	Permeate	0	0	0	0	0	0	0	0
Dibenzo(a,h)anthracene	Influent	0	0	0	0	0	0	0	0
Dibenzo(a,h)anthracene	Permeate	0	0	0	0	0	0	0	0
Benzo(g,h,i)perylene	Influent	0	0	0	0	0.044	0	0.018	0
Benzo(g,h,i)perylene	Permeate	0	0	0	0	0	0	0	0
Chrysene	Influent	0.115	0.052	0.177	0.061	0.541	0.124	0.185	0.144
Chrysene	Permeate	0	0	0	0	0	0	0.017	0

4.2.5. Membrane Fouling Analysis

The impact of operating parameters on the membrane fouling is shown in Figure 4.6 (a, b). Results showed that pore blocking resistance was the most dominant resistance based on RIS model in this study due to the characteristics of oily seawater. Figure 4.6 (a, b) shows that runs with high aeration flow rate and low membrane flux were more effective in maintaining low total resistance (i.e., consisting of cake and pore blocking resistances, and intrinsic membrane resistance).

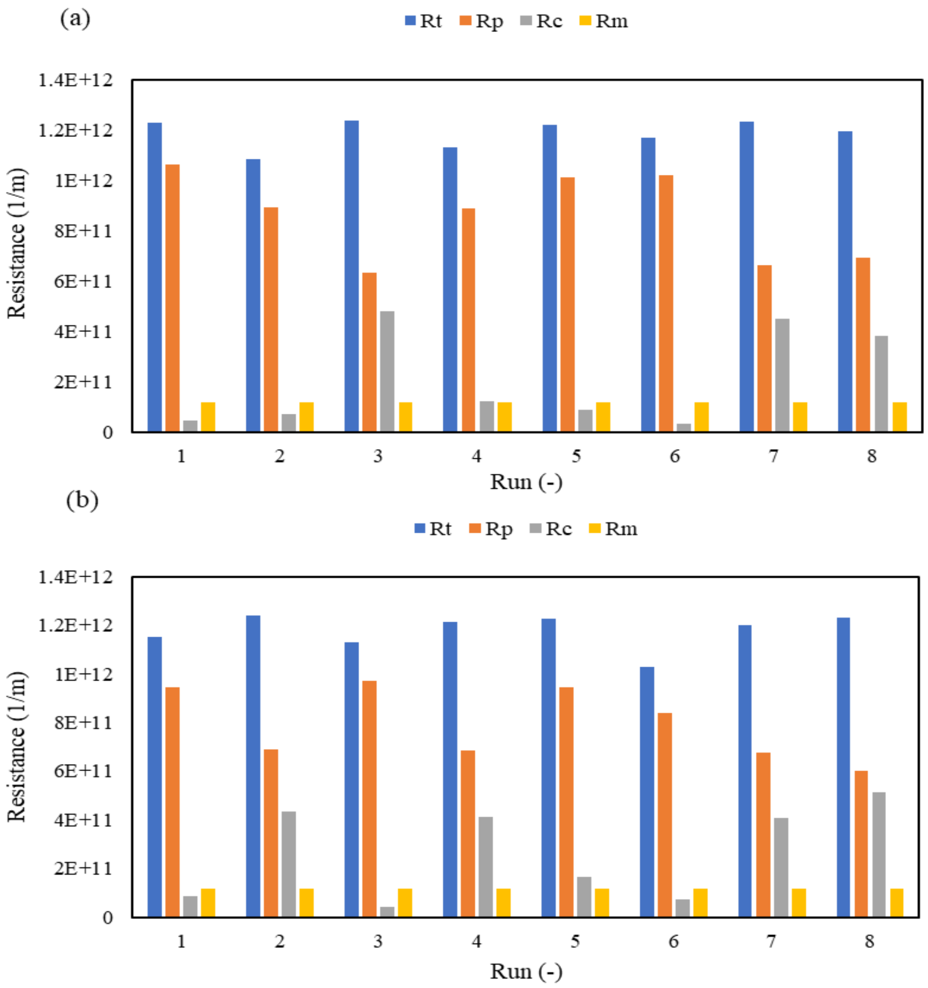

Figure 4.6 Membrane resistances in experiments with (a) CLD, (b) VLSFO

In the runs with lower cake resistance, pore blocking resistance was higher due to the decline of cake thickness which was consistent with the results obtained by Keyvan Hosseini et al. (2020). The amount of cake resistance was not significant due to using aeration and high shear rate inside the membrane tank which dislodged the accumulated layer on the membrane surface similar to the results presented by Braak et al. (2011). This was also in line with Ueda's et al. (1997) results, who investigated the impact of aeration on cake-removing efficiency in a submerged membrane filtration system equipped with HF membranes. Using low oil concentration did not lead to significant accumulation of oily substances on the membrane surface which was consistent with Mohammadi's et al. (2003) study. They showed that higher oil concentration led permeation flux to have higher reduction at a constant TMP and this was because gel resistance increased linearly with increasing oil concentration.

Short operational time, low oil concentration (less than 100 ppm), and using aeration and hydrophilic membrane with antifouling features did not lead to a significant membrane fouling phenomenon. In submerged HF membrane filtration systems, the bubbles generated from aeration create unsteady-state shear at the membrane surface through turbulent eddies, fiber oscillations, particle scouring, and recirculation of content in the membrane tank which lead to the cake layer formation reduction on the membrane surface (Akhondi et al., 2017; Bérubé, 2020; Soydemir et al., 2020).

The regression model of total resistance for CLD and VLSFO (Eqs. (4.3) and (4.4)) showed that R^2 was 81.58% and 68.54%, respectively which 18.42% and 31.46% of the total variance could not be explained by the models for CLD and VLSFO, respectively. In CLD, aeration flow rate was the most significant factor affecting the total resistance (p-value <0.05). However, none of the parameters has a significant impact on the total resistance in VLSFO experiments (p-value>0.05) (as shown in Table 4.5). This would be as a result of the insignificant difference between the selected aeration flow rates in VLSFO runs which led to a relatively similar condition in the experiments. The physical features of two types of oil may affect the experimental runs as well. VLSFO has lower density and viscosity and more volatile chemical compounds compared to CLD which could be another factor.

$$R_t = 1.478\times10^{12} + 3.633\times10^{9} Q_{membrane} - 6.958\times10^{10} Q_{air} + 6.834\times10^{8} C_{oil} \tag{4.3}$$

$$R_t = 1.444\times10^{12} + 1.448\times10^{10} Q_{membrane} - 8.594\times10^{10} Q_{air} - 2.552\times10^{8} C_{oil} \tag{4.4}$$

where C_{oil} refers to oil concentration (ppm), $Q_{membrane}$ is the membrane flux (L/m^2·h), and Q_{air} is the aeration flow rate (m^3/h).

Table 4.5 ANOVA results for total resistance of CLD and VLSFO

Total Resistance of CLD

Source	**Degree of Freedom**	**F-Value**	**P-Value**
Regression	3	5.91	0.06
Oil concentration	1	2.4	0.196
Membrane flux	1	0.98	0.379
Aeration flow rate	1	14.34	0.019
Error	4	-	-
Total	7	-	-

Total Resistance of VLSFO

Source	**Degree of Freedom**	**F-Value**	**P-Value**
Regression	3	2.91	0.165
Oil concentration	1	0.11	0.753

Membrane flux	1	5.29	0.083
Aeration flow rate	1	3.31	0.143
Error	4	-	-
Total	7	-	-

CHAPTER 5 CONCLUSION

5.1. Summary and Conclusion

In this study, comprehensive research on the performance of a pilot-scale membrane filtration system to treat synthetic oily seawater was conducted to recognize its potentials for safe decanting and efficacious treatment. The first stage of this research was to simulate a homogenous and stable oily seawater using a high shear vertical mixer. The synthetic oily seawater was analyzed in terms of oil droplet size, salinity, TDS, conductivity, and viscosity. Oil droplet size was in the range of 1 to 10 μm showing the stability of the synthetic oily wastewater.

The experiments were performed using UF membranes with CLD and VLSFO considering two operating parameters such as aeration flow rate and membrane flux. High TPH removal efficiency was achieved when low membrane flux (6 $L/m^{2}\cdot h$) and moderate aeration flow rate (4.8 m^3/h) were applied to the system. The system met MARPOL 73/78 regulation and decreased the oil concentration in the permeate significantly and the TPH removal efficiency was higher than 91%. The analysis of PHC fractions were conducted and the percentage removal of F3 fraction in all runs was the highest, followed by F2, F1, and F4, respectively. PAH compounds decreased significantly showing the capability of UF membrane to mitigate toxic substances. Consequently, the pilot-scale membrane filtration system with UF membranes was proven to be appropriate as an onsite treatment technology in oil spill response operation.

The investigation of membrane fouling was also conducted through examining fouling resistances. In the experiments, the use of hydrophilic membrane, low oil concentration (less then 100 ppm), proper cleaning methods, and aeration led to an insignificant fouling phenomenon. The biggest fraction of total resistance was related to pore blocking resistance based on RIS model. It was observed that low membrane flux (6 $L/m^2 \cdot h$) and high aeration flow rate (6 m^3/h) resulted in low total resistance in the experiments.

Membrane filtration technology is an appropriate technique by freeing the storage space that is occupied by the recovered water and meets regulatory requirements in order to discharge back the decanted water into the ocean. Therefore, this technology will significantly reduce the number of frequent trips to shore for waste disposal, save time and resources, and increase the overall efficiency and capacity of oil spill response operation. The findings provide scientific aid for decision-making and help mitigate the negative impacts of spilled oil in the ocean.

5.2. Recommendations for Future Research

The following recommendations provide suggestions for further studies:

- Additional research into the capability of the system in treating weathered and fresh crude oils can be conducted.
- A long-term evaluation of the membrane filtration system could be helpful in approving the efficiency of the system both in treatment and membrane fouling.
- The effect of changing time intervals and frequency in membrane cleaning methods such as relaxation and backwashing can be considered.
- Using intermittent aeration can be investigated to save energy consumption and make the system more sustainable.

- Environmentally-friendly chemicals can be used instead of regular acid and alkaline solutions for chemical cleaning to make the pilot-scale membrane filtration system more ecofriendly.
- The addition of a pre-treatment method can help increase initial oil concentration at the beginning of the treatment process without significant membrane fouling and operational failure.

REFERENCES

Abdel-Karim, A., Gad-Allah, T. A., El-Kalliny, A. S., Ahmed, S. I. A., Souaya, E. R., Badawy, M. I., & Ulbricht, M. (2017). Fabrication of modified polyethersulfone membranes for wastewater treatment by submerged membrane bioreactor. Separation and Purification Technology, 175, 36–46. https://doi.org/10.1016/j.seppur.2016.10.060

Abdelrasoul, A. (2015). Investigation on membrane fouling in ultrafiltration of latex solution. PhD thesis, Ryerson University https://rshare.library.ryerson.ca/articles/thesis/Investigation_on_membrane_fouling_in_ultrafiltration_of_latex_solution/14654874

Abidli, A., Huang, Y., Cherukupally, P., Bilton, A. M., & Park, C. B. (2020). Novel separator skimmer for oil spill cleanup and oily wastewater treatment: From conceptual system design to the first pilot-scale prototype development. Environmental Technology & Innovation, 18, 100598. https://doi.org/10.1016/j.eti.2019.100598

Abuhasel, K., Kchaou, M., Alquraish, M., Munusamy, Y., & Jeng, Y. T. (2021). Oily Wastewater Treatment: Overview of Conventional and Modern Methods, Challenges, and Future Opportunities. Water, 13(7), 980. https://doi.org/10.3390/w13070980

Adetunji, A. I., & Olaniran, A. O. (2021). Treatment of industrial oily wastewater by advanced technologies: A review. Applied Water Science, 11(6), 98. https://doi.org/10.1007/s13201-021-01430-4

Ahamed, M.I.; Lichtfouse, E. (Eds.) (2021) Water Pollution and Remediation: Organic Pollutants; Springer International Publishing, ISBN 978-3-030-52394-7

Ahmad, A. L., Bhatia, S., Ibrahim, N., & Sumathi, S. (2005). Adsorption of residual oil from palm oil mill effluent using rubber powder. Brazilian Journal of Chemical Engineering, 22(3), 371–379. https://doi.org/10.1590/S0104-66322005000300006

Ahmed, I., Balkhair, K. S., Albeiruttye, M. H., & Shaiban, A. A. J. (2017). Importance and significance of UF/MF membrane systems in desalination water treatment. Desalination, 187. http://dx.doi.org/10.5772/intechopen.68694

Akhondi, E., Zamani, F., Tng, K., Leslie, G., Krantz, W., Fane, A., & Chew, J. (2017). The Performance and Fouling Control of Submerged Hollow Fiber (HF) Systems: A Review. Applied Sciences, 7(8), 765. https://doi.org/10.3390/app7080765

Aliff Radzuan, M. R., Abia-Biteo Belope, M. A., & Thorpe, R. B. (2016). Removal of fine oil droplets from oil-in-water mixtures by dissolved air flotation. Chemical Engineering Research and Design, 115, 19–33. https://doi.org/10.1016/j.cherd.2016.09.013

Al-Khafaji, S. S., Al-Rekabi, W. S., & Mawat, M. J. (2022). Apply Membrane Biological Reactor (MBR) in Industrial wastewater treatment: A Mini Review. Eurasian Journal of Engineering and Technology, 7, 98–106. https://www.geniusjournals.org/index.php/ejet/article/view/1804

Altinbas, M., Ozturk, H., & İren, E. (2021). Full Scale Sanitary Landfill Leachate Treatment by MBR: Flat Sheet vs. Hollow Fiber Membrane. Journal of Membrane Science and Research, 7(2), 118–124. https://doi.org/10.22079/jmsr.2020.123563.1358

Alzahrani, S., & Mohammad, A. W. (2014). Challenges and trends in membrane technology implementation for produced water treatment: A review. Journal of Water Process Engineering, 4, 107–133. https://doi.org/10.1016/j.jwpe.2014.09.007

Anis, S. F., Hashaikeh, R., & Hilal, N. (2019). Microfiltration membrane processes: A review of research trends over the past decade. Journal of Water Process Engineering, 32, 100941. https://doi.org/10.1016/j.jwpe.2019.100941

Avery, D. (2020). Death by a thousand spills: How corporate branding and media strategies downplay the risk of offshore oil spills in Canada. Journal of Environmental Media, 1(2), 225–244. https://doi.org/10.1386/jem_00017_1

Ayub, M., Othman, M. H. D., Kadir, S. H. S. A., Ali, A., Khan, I. U., Yusop, M. Z. M., Matsuura, T., Fauzi Ismail, A., A. Rahman, M., & Jaafar, J. (2021). Research and Development Journey and Future Trends of Hollow Fiber Membranes for Purification Applications (1970–2020): A Bibliometric Analysis. Membranes, 11(8), 600. https://doi.org/10.3390/membranes11080600

Badrnezhad, R., & Beni, A. H. (2013). Ultrafiltration membrane process for produced water treatment: Experimental and modeling. Journal of Water Reuse and Desalination, 3(3), 249–259. https://doi.org/10.2166/wrd.2013.092

Bagheri, M., & Mirbagheri, S. A. (2018). Critical review of fouling mitigation strategies in membrane bioreactors treating water and wastewater. Bioresource Technology, 258, 318–334. https://doi.org/10.1016/j.biortech.2018.03.026

Baker, R. W. (2004). Membrane Technology and Applications. John Wiley & Sons. https://doi.org/10.1002/0470020393

Baker, R. W. (2012). Membrane technology and applications. John Wiley & Sons.

Bakke, T., Klungsøyr, J., & Sanni, S. (2013). Environmental impacts of produced water and drilling waste discharges from the Norwegian offshore petroleum industry. Marine Environmental Research, 92, 154–169. https://doi.org/10.1016/j.marenvres.2013.09.012

Barjoveanu, G., & Teodosiu, C. (2006). Advanced treatment for pulp and paper wastewater recycling by membrane processes. Environmental Engineering & Management Journal (EEMJ), 5(2). http://dx.doi.org/10.30638/eemj.2006.012

Bechhold, H. (1907). Kolloidstudien mit der Filtrationsmethode. Zeitschrift für Physikalische Chemie, 60U(1), 257–318. https://doi.org/10.1515/zpch-1907-6013

Berk, Z. (2018). Food Process Engineering and Technology. Academic Press.

Bérubé, P. R. (2020). 11-Air sparging for fouling control. In Current Developments in Biotechnology and Bioengineering (pp. 225–243). Elsevier. https://doi.org/10.1016/B978-0-12-819809-4.00011-5

Beyer, J., Trannum, H. C., Bakke, T., Hodson, P. V., & Collier, T. K. (2016). Environmental effects of the Deepwater Horizon oil spill: A review. Marine Pollution Bulletin, 110(1), 28–51. https://doi.org/10.1016/j.marpolbul.2016.06.027

Bhattacharjee, S., Kim, A. S., & Elimelech, M. (1999). Concentration Polarization of Interacting Solute Particles in Cross-Flow Membrane Filtration. Journal of Colloid and Interface Science, 212(1), 81–99. https://doi.org/10.1006/jcis.1998.6045

Blandin, G., Verliefde, A. R. D., Comas, J., Rodriguez-Roda, I., & Le-Clech, P. (2016). Efficiently Combining Water Reuse and Desalination through Forward Osmosis-Reverse Osmosis (FO-RO) Hybrids: A Critical Review. Membranes, 6(3), 37. https://doi.org/10.3390/membranes6030037

Bonisoli-Alquati, A., Stouffer, P. C., Turner, R. E., Woltmann, S., & Taylor, S. S. (2016). Incorporation of Deepwater Horizon oil in a terrestrial bird. Environmental Research Letters, 11(11), 114023. https://doi.org/10.1088/1748-9326/11/11/114023

Bopape, M. F., Van Geel, T., Dutta, A., Van der Bruggen, B., & Onyango, M. S. (2021). Numerical Modelling Assisted Design of a Compact Ultrafiltration (UF) Flat Sheet Membrane Module. Membranes, 11(1), 54. https://doi.org/10.3390/membranes11010054

Braak, E., Alliet, M., Schetrite, S., & Albasi, C. (2011). Aeration and hydrodynamics in submerged membrane bioreactors. Journal of Membrane Science, 379(1–2), 1–18. https://doi.org/10.1016/j.memsci.2011.06.004

Buer, T., & Cumin, J. (2010). MBR module design and operation. Desalination, 250(3), 1073–1077. https://doi.org/10.1016/j.desal.2009.09.111

Campo, R., Giustra, M., Marchis, M., Freni, G., & Bella, G. (2017). Characterization and Treatment Proposals of Shipboard Slop Wastewater Contaminated by Hydrocarbons. Water, 9(8), 581. https://doi.org/10.3390/w9080581

Cao, Y. (2016). Examinations of the shear stress on MBR-membrane plates by a single bubble using CFD. Master's Thesis, Technische Universität. https://tuprints.ulb.tu-darmstadt.de/5273/

CBC. (2019). Cargo ship acquitted of charges relating to 2015 oil spill. Available online: https://www.cbc.ca/news/canada/british-columbia/marathassa-acquitted-charges-2015-oil-spill-1.5010021 (accessed on 16 September 2022)

CBC. (2021). Husky Energy facing 3 charges for massive SeaRose oil spill off coast of N.L. Available online: https://www.cbc.ca/news/canada/newfoundland-labrador/oil-spill-newfoundland-husky-charges-1.6216670 (accessed on 16 September 2022)

Chakrabarty, B., Ghoshal, A. K., & Purkait, M. K. (2008). Ultrafiltration of stable oil-in-water emulsion by polysulfone membrane. Journal of Membrane Science, 325(1), 427–437. https://doi.org/10.1016/j.memsci.2008.08.007

ChemSafetyPro. (2021). Water Solubility. Available online: https://www.chemsafetypro.com/Topics/CRA/Water_Solubility.html (accessed on 25 July 2022)

Chen, B., Liu, B., Ling, J., Matchinski, E. J., Dong, G., Ye, X., Wu, F., Shen, W., Li, J., Liu, L., Lee, K., Isaacman, L., Potter, S., Hynes, B., & Zhang, B. (2022). Development of Advanced Oil/Water Separation Technologies to Enhance the Effectiveness of Mechanical Oil Recovery Operations at Sea: Potential and Challenges, Journal of Hazardous Materials, 129340 https://doi.org/10.1016/j.jhazmat.2022.129340

Cheng, B., Li, Z., Li, Q., Ju, J., Kang, W., & Naebe, M. (2017). Development of smart poly(vinylidene fluoride)-graft-poly(acrylic acid) tree-like nanofiber membrane for pH-responsive oil/water separation. Journal of Membrane Science, 534, 1–8. https://doi.org/10.1016/j.memsci.2017.03.053

Cheryan, M., & Rajagopalan, N. (1998). Membrane processing of oily streams. Wastewater treatment and waste reduction. Journal of Membrane Science, 151(1), 13–28. https://doi.org/10.1016/S0376-7388(98)00190-2

Cho, D.O. (2007). The effects of the M/V Sea Prince accident on maritime safety management in Korea. Marine Policy, 31(6), 730–735. https://doi.org/10.1016/j.marpol.2007.01.005

Chua, K. T., Hawlader, M. N. A., & Malek, A. (2003). Pretreatment of seawater: Results of pilot trials in Singapore. Desalination, 159(3), 225–243. https://doi.org/10.1016/S0011-9164(03)90075-0

Conmy, R., Barron, M., SantoDomingo, J., & Deshpande, R. (2017). Characterization and Behavior of Cold Lake Blend and Western Canadian Select Diluted Bitumen Products. United States Environmental Protection Agency. https://nepis.epa.gov/Exe/ZyPDF.cgi/P100TN0P.PDF?Dockey=P100TN0P.PDF

Dalaklis, D., Christodoulou, A., & Kitada, M. (2020). Oil spill response training in the south Baltic Sea region. 3309–3314. https://doi.org/10.21125/inted.2020.0948

Dale, M., Keown, R., & Wilson, G. (2013). Following The Safe Evacuation of All Offshore Personnel, the Environmental Response Takes Primacy: Learning from The Elgin G4 Well Major Incident. SPE Offshore Europe Oil and Gas Conference and Exhibition. https://doi.org/10.2118/166590-MS

Di Bella, G., & Di Trapani, D. (2019). A Brief Review on the Resistance-in-Series Model in Membrane Bioreactors (MBRs). Membranes, 9(2), 24. https://doi.org/10.3390/membranes9020024

Di Profio, G., Ji, X., Curcio, E., & Drioli, E. (2011). Submerged hollow fiber ultrafiltration as seawater pretreatment in the logic of integrated membrane desalination systems. Desalination, 269(1–3), 128–135. https://doi.org/10.1016/j.desal.2010.10.051

Dickhout, J. M., Moreno, J., Biesheuvel, P. M., Boels, L., Lammertink, R. G. H., & de Vos, W. M. (2017). Produced water treatment by membranes: A review from a colloidal perspective. Journal of Colloid and Interface Science, 487, 523–534. https://doi.org/10.1016/j.jcis.2016.10.013

Dickhout, J. M., Kleijn, J. M., Lammertink, R. G. H., & de Vos, W. M. (2018). Adhesion of emulsified oil droplets to hydrophilic and hydrophobic surfaces - effect of surfactant charge, surfactant concentration and ionic strength. Soft Matter, 14(26), 5452–5460. https://doi.org/10.1039/C8SM00476E

Doyen, W., Mues, W., Molenberghs, B., & Cobben, B. (2010). Spacer fabric supported flat-sheet membranes: A new era of flat-sheet membrane technology. Desalination, 250(3), 1078–1082. https://doi.org/10.1016/j.desal.2009.09.112

Drews, A. (2010). Membrane fouling in membrane bioreactors—Characterisation, contradictions, cause and cures. Journal of Membrane Science, 363(1–2), 1–28. https://doi.org/10.1016/j.memsci.2010.06.046

Du, X., Shi, Y., Jegatheesan, V., & Haq, I. U. (2020). A Review on the Mechanism, Impacts and Control Methods of Membrane Fouling in MBR System. Membranes, 10(2), 24. https://doi.org/10.3390/membranes10020024

El-badawy, T., Othman, M. H. D., Adam, M. R., Kamaludin, R., Ismail, A. F., Rahman, M. A., Jaafar, J., Rajabzadeh, S., Matsuyama, H., Usman, J., Kurniawan, T. A., Raji, Y. O., Mamah, S. C., & Abdullah, H. (2022). Braid-reinforced PVDF hollow fiber membranes for high-efficiency separation of oily wastewater. Journal of Environmental Chemical Engineering, 10(2), 107258. https://doi.org/10.1016/j.jece.2022.107258

Elford, W. J., Grabar, P., & Ferry, J. D. (1935). Graded Collodion Membranes for Bacteriological Studies. Practical Aspects of the Mechanism Determining the Character of the Membrane, and the Roles of Particular Solvent Constituents. British Journal of Experimental Pathology, 16(6), 583–587. https://www.ncbi.nlm.nih.gov/pmc/articles/PMC2065226/

Etemadi, H., Fonouni, M., & Yegani, R. (2020). Investigation of antifouling properties of polypropylene/TiO_2 nanocomposite membrane under different aeration rate in membrane bioreactor system. Biotechnology Reports, 25, e00414. https://doi.org/10.1016/j.btre.2019.e00414

Feng, Q., An, C. J., Cao, Y., Chen, Z., Owens, E., Taylor, E., Wang, Z., & Saad, E. A. (2021). An Analysis of Selected Oil Spill Case Studies on the Shorelines of Canada. Journal of Environmental Informatics Letters, 5(1), 39-47.5(1), 39–47.

Fingas, M. F., & Brown, C. E. (1997). Review of oil spill remote sensing. Spill Science & Technology Bulletin, 4(4), 199-208. https://doi.org/10.1016/S1353-2561(98)00023-1

Fingas, M. F. (1999). The Evaporation of Oil Spills: Development and Implementation of New Prediction Methodology. International Oil Spill Conference Proceedings, 1999(1), 281–287. https://doi.org/10.7901/2169-3358-1999-1-281

Fingas, M. (2016). Oil spill science and technology. Gulf professional publishing. https://www.elsevier.com/books/oil-spill-science-and-technology/fingas/978-0-12-809413-6

Frederickson, K. C. (2005). The Application of a Membrane Bioreactor for Wastewater Treatment on a Northern Manitoban Aboriginal Community. 126. Master's thesis, University of Manitoba. https://mspace.lib.umanitoba.ca/handle/1993/188

Galieriková, A., & Materna, M. (2020). World Seaborne Trade with Oil: One of Main Cause for Oil Spills? Transportation Research Procedia, 44, 297–304. https://doi.org/10.1016/j.trpro.2020.02.039

Germain, E., Stephenson, T., & Pearce, P. (2005). Biomass characteristics and membrane aeration: Toward a better understanding of membrane fouling in submerged membrane bioreactors (MBRs). Biotechnology and Bioengineering, 90(3), 316–322. https://doi.org/10.1002/bit.20411

Ghidossi, R., Veyret, D., Scotto, J. L., Jalabert, T., & Moulin, P. (2009). Ferry oily wastewater treatment. Separation and Purification Technology, 64(3), 296–303. https://doi.org/10.1016/j.seppur.2008.10.013

Gill, D. A., Picou, J. S., & Ritchie, L. A. (2012). The Exxon Valdez and BP oil spills: A comparison of initial social and psychological impacts. American Behavioral Scientist, 56(1), 3-23. https://doi.org/10.1177%2F0002764211408585

Gkotsis, P., Banti, D., Peleka, E., Zouboulis, A., & Samaras, P. (2014). Fouling Issues in Membrane Bioreactors (MBRs) for Wastewater Treatment: Major Mechanisms, Prevention and Control Strategies. Processes, 2(4), 795–866. https://doi.org/10.3390/pr2040795

Goh, P. S., Ong, C. S., Ng, B. C., & Ismail, A. F. (2019). Applications of emerging nanomaterials for oily wastewater treatment. In Nanotechnology in Water and Wastewater Treatment, 101-113. Elsevier. https://doi.org/10.1016/B978-0-12-813902-8.00005-8

González-Pérez, D. M., Garralón, G., Plaza, F., Pérez, J. I., Moreno, B., & Gómez, M. A. (2012). Removal of low concentrations of phenanthrene, fluoranthene and pyrene from urban wastewater by membrane bioreactors technology. Journal of Environmental Science and Health, Part A, 47(14), 2190–2197. https://doi.org/10.1080/10934529.2012.707496

Government of Canada. (n.d.a). Fact sheet: 2-methylnaphthalene. Available online: https://gost.tpsgc-pwgsc.gc.ca/Contfs.aspx?ID=28&lang=eng (accessed on 8 September 2022)

Government of Canada. (n.d.b). Fact sheet: Phenanthrene. Available online: https://gost.tpsgc-pwgsc.gc.ca/Contfs.aspx?ID=43&lang=eng (accessed on 8 September 2022)

Government of Canada. (2018). Canadian Environmental Protection Act. Available online: https://laws-lois.justice.gc.ca/eng/ acts/c-15.31/page-20.html#docCont (accessed on 20 July 2021).

Gul, A., Hruza, J., & Yalcinkaya, F. (2021). Fouling and Chemical Cleaning of Microfiltration Membranes: A Mini-Review. Polymers, 13(6), 846. https://doi.org/10.3390/polym13060846

Han, G., de Wit, J. S., & Chung, T.S. (2015). Water reclamation from emulsified oily wastewater via effective forward osmosis hollow fiber membranes under the PRO mode. Water Research, 81, 54–63. https://doi.org/10.1016/j.watres.2015.05.048

Han, M., Zhang, J., Chu, W., Chen, J., & Zhou, G. (2019). Research Progress and Prospects of Marine Oily Wastewater Treatment: A Review. Water, 11(12), 2517. https://doi.org/10.3390/w11122517

Hesampour, M., Krzyzaniak, A., & Nyström, M. (2008). The influence of different factors on the stability and ultrafiltration of emulsified oil in water. Journal of Membrane Science, 325(1), 199–208. https://doi.org/10.1016/j.memsci.2008.07.048

Hilal, N., Ogunbiyi, O., Miles, N., & Nigmatullin, R. (2005). Methods Employed for Control of Fouling in MF and UF Membranes: A Comprehensive Review. https://doi.org/10.1081/SS-200068409

Hodson, P. V. (2017). The toxicity to fish embryos of PAH in crude and refined oils. Archives of environmental contamination and toxicology, 73(1), 12-18. https://doi.org/10.1007/s00244-016-0357-6

Hong, A., Fane, A. G., & Burford, R. (2003). Factors affecting membrane coalescence of stable oil-in-water emulsions. Journal of Membrane Science, 222(1), 19–39. https://doi.org/10.1016/S0376-7388(03)00137-6

Huang, Q., & Long, X. (2020). Analysis of the Influencing Factors on Oil Removal Efficiency in Large-Scale Flotation Tanks: Experimental Observation and Numerical Simulation. Energies, 13(4), 927. https://doi.org/10.3390/en13040927

Huang, S., Ras, R. H. A., & Tian, X. (2018). Antifouling membranes for oily wastewater treatment: Interplay between wetting and membrane fouling. Current Opinion in Colloid & Interface Science, 36, 90–109. https://doi.org/10.1016/j.cocis.2018.02.002

Huang, X., Wang, W., Liu, Y., Wang, H., Zhang, Z., Fan, W., & Li, L. (2015). Treatment of oily waste water by PVP grafted PVDF ultrafiltration membranes. Chemical Engineering Journal, 273, 421–429. https://doi.org/10.1016/j.cej.2015.03.086

Huang, Z., Clark, W. R., & Ronco, C. (2012). Convective Clearance. In J.L. Vincent & J. B. Hall (Eds.), Encyclopedia of Intensive Care Medicine (pp. 611–614). Springer. https://doi.org/10.1007/978-3-642-00418-6_266

Hube, S., Eskafi, M., Hrafnkelsdóttir, K. F., Bjarnadóttir, B., Bjarnadóttir, M. Á., Axelsdóttir, S., & Wu, B. (2020). Direct membrane filtration for wastewater treatment and resource recovery: A review. Science of The Total Environment, 710, 136375. https://doi.org/10.1016/j.scitotenv.2019.136375

Hussain, M. S. (2019). Modelling and prediction of non-linear scale-up from an Ultra Scale-Down membrane device to process scale tangential flow filtration, Doctoral thesis, UCL (University College London). https://discovery.ucl.ac.uk/id/eprint/10078635/

Inchem. (2006). Acenaphthene. Available online: https://inchem.org/documents/icsc/icsc/eics1674.htm (accessed on 4 September 2022)

Inoxpa. (2021). ME1100 Vertical Mixer. Available online: https://www.inoxpa.com/products/mixing/high-shear-mixers/vertical-mixer (accessed on 19 December 2021)

Ipieca. (2013). The use of decanting during offshore oil spill recovery operations. Available online: https://www.ipieca.org/resources/awareness-briefing/the-use-of-decanting-during-offshore-oil-spill-recovery-operations (accessed on 24 April 2022)

ISCO. (2022). Guidelines for decanting settled out water. Available online: https://spillcontrol.org/2015/03/02/guidelines-for-decanting-settled-out-water (accessed on 9 April 2022)

Ismail, N. H., Salleh, W. N. W., Ismail, A. F., Hasbullah, H., Yusof, N., Aziz, F., & Jaafar, J. (2020). Hydrophilic polymer-based membrane for oily wastewater treatment: A review. Separation and Purification Technology, 233, 116007. https://doi.org/10.1016/j.seppur.2019.116007

ITOPF. (2012). Use of Skimmers in Oil Pollution Response. Available online: https://www.itopf.org/fileadmin/uploads/itopf/data/Documents/TIPS_TAPS_new/TIP_5_Use_of_Skimmers_in_Oil_Pollution_Response.pdf (accessed on 6 August 2022)

Izadi, A., Hosseini, M., Najafpour Darzi, G., Nabi Bidhendi, G., Pajoum Shariati, F., & Mosaddeghi, M. R. (2018). Perspectives on Membrane Bioreactor Potential for Treatment of Pulp and Paper Industry Wastewater: A Critical Review. Journal of Applied Biotechnology Reports, 5(4), 139–150. https://doi.org/10.29252/JABR.05.04.02

Jamaly, S., Darwish, N. N., Ahmed, I., & Hasan, S. W. (2014). A short review on reverse osmosis pretreatment technologies. Desalination, 354, 30–38. https://doi.org/10.1016/j.desal.2014.09.017

Johari, A., Razmjouei, M., Mansourizadeh, A., & Emadzadeh, D. (2020). Fabrication of blend hydrophilic polyamide imide (Torlon®)-sulfonated poly (ether ether ketone) hollow fiber membranes for oily wastewater treatment. Polymer Testing, 91, 106733. https://doi.org/10.1016/j.polymertesting.2020.106733

Judd, S. (2005). Fouling control in submerged membrane bioreactors. Water Science and Technology : A Journal of the International Association on Water Pollution Research, 51, 27–34. https://doi.org/10.2166/wst.2005.0618

Judd, S. (2010). The MBR book: Principles and applications of membrane bioreactors for water and wastewater treatment.

Judd, S. J. (2016). The status of industrial and municipal effluent treatment with membrane bioreactor technology. Chemical Engineering Journal, 305, 37–45. https://doi.org/10.1016/j.cej.2015.08.141

Judd, S., & Turan, F. (2018). Sidestream vs immersed membrane bioreactors: A cost analysis. Proceedings of the Water Environment Federation, 2018, 3722–3733. https://doi.org/10.2175/193864718825136008

Kang, Y., Xia, Y., Wang, H., & Zhang, X. (2019). 2D Laminar Membranes for Selective Water and Ion Transport. Advanced Functional Materials, 29(29), 1902014. https://doi.org/10.1002/adfm.201902014

Karakulski, K., & Gryta, M. (2017). The application of ultrafiltration for treatment of ships generated oily wastewater. Chemical Papers, 71(6), 1165–1173. https://doi.org/10.1007/s11696-016-0108-1

Keyvan Hosseini, M., Liu, L., Keyvan Hosseini, P., Bhattacharyya, A., Lee, K., Miao, J., & Chen, B. (2022). Review of Hollow Fiber (HF) Membrane Filtration Technology for the Treatment of Oily Wastewater: Applications and Challenges. Journal of Marine Science and Engineering, 10(9), 1313. https://doi.org/10.3390/jmse10091313

Keyvan Hosseini, P., Pajoum Shariati, F., Delavari Amrei, H., & Heydarinasab, A. (2020). The influence of various orifice diameters on cake resistance and pore blocking resistance of a hybrid membrane photobioreactor (HMPBR). Separation and Purification Technology, 235, 116187. https://doi.org/10.1016/j.seppur.2019.116187

Keyvan Hosseini, P., Liu, L., Keyvan Hosseini, M., Bhattacharyya, A., Miao, J., & Wang, F. (2023). Treatment of a synthetic decanted oily seawater in a pilot-scale hollow fiber membrane filtration process: Experimental investigation. Journal of Hazardous Materials, 441, 129928. https://doi.org/10.1016/j.jhazmat.2022.129928

Kharraz, J. A., Khanzada, N. K., Farid, M. U., Kim, J., Jeong, S., & An, A. K. (2022). Membrane distillation bioreactor (MDBR) for wastewater treatment, water reuse, and resource recovery: A review. Journal of Water Process Engineering, 47, 102687. https://doi.org/10.1016/j.jwpe.2022.102687

Kim, B. r., Kalis, E. m., Florkey, D. l., Swatsenbarg, S. l., Luciw, L., Bailey, C. h., Gaines, W. a., Phillips, J. h., & Kosokowsky, G. b. (1998). Evaluation of commercial ultrafiltration systems for treating automotive oily wastewater. Water Environment Research, 70(7), 1280–1289. https://doi.org/10.2175/106143098X123651

Kim, D., Seo, J., Ahn, S., & Lee, H. (2021). Effectiveness of Dispersants for Very-Low-Sulfur Fuel Oil. Journal of the Korean Society of Marine Environment and Safety, 27(1), 113–118. https://doi.org/10.7837/kosomes.2021.27.1.113

Kim, K. D., Choi, D. W., Choa, Y.-H., & Kim, H. T. (2007). Optimization of parameters for the synthesis of zinc oxide nanoparticles by Taguchi robust design method. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 311(1–3), 170–173. https://doi.org/10.1016/j.colsurfa.2007.06.017

Kose, B., Ozgun, H., Ersahin, M. E., Dizge, N., Koseoglu-Imer, D. Y., Atay, B., Kaya, R., Altınbas, M., Sayılı, S., Hoshan, P., Atay, D., Eren, E., Kinaci, C., & Koyuncu, I. (2012). Performance evaluation of a submerged membrane bioreactor for the treatment of brackish oil and natural gas field produced water. Desalination, 285, 295–300. https://doi.org/10.1016/j.desal.2011.10.016

Kraume, M., Wedi, D., Schaller, J., Iversen, V., & Drews, A. (2009). Fouling in MBR: What use are lab investigations for full scale operation? Desalination, 236(1), 94–103. https://doi.org/10.1016/j.desal.2007.10.055

Krzeminski, P., Leverette, L., Malamis, S., & Katsou, E. (2017). Membrane bioreactors – A review on recent developments in energy reduction, fouling control, novel configurations, LCA and market prospects. Journal of Membrane Science, 527, 207–227. https://doi.org/10.1016/j.memsci.2016.12.010

Kuyukina, M. S., Krivoruchko, A. V., & Ivshina, I. B. (2020). Advanced Bioreactor Treatments of Hydrocarbon-Containing Wastewater. Applied Sciences, 10(3), 831. https://doi.org/10.3390/app10030831

Le, T. V., Imai, T., Higuchi, T., Yamamoto, K., Sekine, M., Doi, R., Vo, H. T., & Wei, J. (2013). Performance of tiny microbubbles enhanced with “normal cyclone bubbles” in separation of fine oil-in-water emulsions. Chemical Engineering Science, 94, 1–6. https://doi.org/10.1016/j.ces.2013.02.044

Le-Clech, P., Jefferson, B., & Judd, S. J. (2005). A comparison of submerged and sidestream tubular membrane bioreactor configurations. Desalination, 173(2), 113–122. https://doi.org/10.1016/j.desal.2004.08.029

Le-Clech, P., Chen, V., & Fane, T. A. G. (2006). Fouling in membrane bioreactors used in wastewater treatment. Journal of Membrane Science, 284(1–2), 17–53. https://doi.org/10.1016/j.memsci.2006.08.019

Lee, K., Boufadel, M., Chen, B., Foght, J., Hodson, P., Swanson, S., & Venosa, A. (2015). Royal Society of Canada Expert Panel Report: The Behaviour and Environmental Impacts of Crude Oil Released into Aqueous Environments. Royal Society of Canada. http://deslibris.ca/ID/10049082

Lenntech. (2022). MBR Introduction. Available online: https://www.lenntech.com/processes/mbr-introduction.htm#ixzz7ZRJteBhB (accessed on 24 May 2022)

Li, C., Yang, Y., Ding, S., & Hou, L.A. (2016). Dynamics of biofouling development on the conditioned membrane and its relationship with membrane performance. Journal of Membrane Science, 514, 264–273. https://doi.org/10.1016/j.memsci.2016.04.066

Li, J., Jiang, C., Shi, W., Song, F., He, D., Miao, H., wang, T., Deng, J., & Ruan, W. (2018). Polytetrafluoroethylene (PTFE) hollow fiber AnMBR performance in the treatment of organic wastewater with varying salinity and membrane cleaning behavior. Bioresource Technology, 267, 363–370. https://doi.org/10.1016/j.biortech.2018.07.063

Li, P., Cai, Q., Lin, W., Chen, B., & Zhang, B. (2016). Offshore oil spill response practices and emerging challenges. Marine Pollution Bulletin, 110(1), 6–27. https://doi.org/10.1016/j.marpolbul.2016.06.020

Li, Q., & Elimelech, M. (2004). Organic Fouling and Chemical Cleaning of Nanofiltration Membranes: Measurements and Mechanisms. Environmental Science & Technology, 38(17), 4683–4693. https://doi.org/10.1021/es0354162

Li, T., Law, A. W.K., & Fane, A. G. (2014). Submerged hollow fibre membrane filtration with transverse and longitudinal vibrations. Journal of Membrane Science, 455, 83–91. https://doi.org/10.1016/j.memsci.2013.12.042

Li, X., Mo, Y., Li, J., Guo, W., & Ngo, H. H. (2017). In-situ monitoring techniques for membrane fouling and local filtration characteristics in hollow fiber membrane processes: A critical review. Journal of Membrane Science, 528, 187–200. https://doi.org/10.1016/j.memsci.2017.01.030

Li, Y. S., Yan, L., Xiang, C. B., & Hong, L. J. (2006). Treatment of oily wastewater by organic–inorganic composite tubular ultrafiltration (UF) membranes. Desalination, 196(1–3), 76–83. https://doi.org/10.1016/j.desal.2005.11.021

Li, Y., Zhang, Z., Ge, B., Men, X., & Xue, Q. (2017). A versatile and efficient approach to separate both surfactant-stabilized water-in-oil and oil-in-water emulsions. Separation and Purification Technology, 176, 1–7. https://doi.org/10.1016/j.seppur.2016.11.072

Li, Z., Lee, K., King, T., Kepkay, P., Boufadel, M. C., & Venosa, A. D. (2009). Evaluating Chemical Dispersant Efficacy in an Experimental Wave Tank: 1, Dispersant Effectiveness as a Function of Energy Dissipation Rate. Environmental Engineering Science, 26(6), 1139–1148. https://doi.org/10.1089/ees.2008.0377

Lin, H., Gao, W., Meng, F., Liao, B.Q., Leung, K.T., Zhao, L., Chen, J., & Hong, H. (2012). Membrane Bioreactors for Industrial Wastewater Treatment: A Critical Review. Critical Reviews in Environmental Science and Technology, 42(7), 677–740. https://doi.org/10.1080/10643389.2010.526494

Liu, Y., Su, Y., Cao, J., Guan, J., Zhang, R., He, M., Fan, L., Zhang, Q., & Jiang, Z. (2017). Antifouling, high-flux oil/water separation carbon nanotube membranes by polymer-mediated surface charging and hydrophilization. Journal of Membrane Science, 542, 254–263. https://doi.org/10.1016/j.memsci.2017.08.018

Lu, D., Cheng, W., Zhang, T., Lu, X., Liu, Q., Jiang, J., & Ma, J. (2016). Hydrophilic Fe2O3 dynamic membrane mitigating fouling of support ceramic membrane in ultrafiltration of oil/water emulsion. Separation and Purification Technology, 165, 1–9. https://doi.org/10.1016/j.seppur.2016.03.034

Luis, P. (2018). Fundamental Modeling of Membrane Systems: Membrane and Process Performance. Elsevier.

Mahbouba, Z. N., Mahmood, A. K., & Alshammari, M. H. (2021). Ultrafiltration Membrane Technology for Oily Wastewater Treatment. IOP Conference Series: Earth and Environmental Science, 877(1), 012012. https://doi.org/10.1088/1755-1315/877/1/012012

Marchese, J., Ochoa, N. A., Pagliero, C., & Almandoz, C. (2000). Pilot-Scale Ultrafiltration of an Emulsified Oil Wastewater. Environmental Science & Technology, 34(14), 2990–2996. https://doi.org/10.1021/es9909069

Marrot, B., Barrios-Martinez, A., Moulin, P., & Roche, N. (2004). Industrial wastewater treatment in a membrane bioreactor: A review. Environmental Progress, 23(1), 59–68. https://doi.org/10.1002/ep.10001

Martini, S., Ang, H. M., & Znad, H. (2017). Integrated Ultrafiltration Membrane Unit for Efficient Petroleum Refinery Effluent Treatment: Water. Clean - Soil, Air, Water, 45(2), 1600342. https://doi.org/10.1002/clen.201600342

Masoudnia, K., Raisi, A., Aroujalian, A., & Fathizadeh, M. (2014). A hybrid microfiltration/ultrafiltration membrane process for treatment of oily wastewater. Desalination and Water Treatment, 55(4), 901-912. https://doi.org/10.1080/19443994.2014.922501

Maximous, N., Nakhla, G., & Wan, W. (2009). Comparative assessment of hydrophobic and hydrophilic membrane fouling in wastewater applications. Journal of Membrane Science, 339(1–2), 93–99. https://doi.org/10.1016/j.memsci.2009.04.034

Maxxam. (2021). Evaluation of the rationale behind new regulation 153 groundwater standards for F3 and F4 petroleum hydrocarbons. available online: https://kodiak.clientszone.net/wp-content/uploads/2021/01/Solubility-for-F3-F4.pdf

McCabe, W. L., Smith, J. C., & Harriott, P. (1993). Unit Operations of Chemical Engineering. McGraw-Hill, New York.

Medeiros, A. D. M. de, Silva Junior, C. J. G. da, Amorim, J. D. P. de, Durval, I. J. B., Costa, A. F. de S., & Sarubbo, L. A. (2022). Oily Wastewater Treatment: Methods, Challenges, and Trends. Processes, 10(4), 743. https://doi.org/10.3390/pr10040743

Melbiah, J. S. B., Nithya, D., & Mohan, D. (2017). Surface modification of polyacrylonitrile ultrafiltration membranes using amphiphilic Pluronic F127/CaCO3 nanoparticles for oil/water emulsion separation. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 516, 147–160. https://doi.org/10.1016/j.colsurfa.2016.12.008

Metzger, U., Le-Clech, P., Stuetz, R. M., Frimmel, F. H., & Chen, V. (2007). Characterisation of polymeric fouling in membrane bioreactors and the effect of different filtration modes. Journal of Membrane Science, 301(1–2), 180–189. https://doi.org/10.1016/j.memsci.2007.06.016

Miller, D. J., Dreyer, D. R., Bielawski, C. W., Paul, D. R., & Freeman, B. D. (2017). Surface Modification of Water Purification Membranes. Angewandte Chemie International Edition, 56(17), 4662–4711. https://doi.org/10.1002/anie.201601509

Mohammadi, L., Rahdar, A., Bazrafshan, E., Dahmardeh, H., Susan, Md. A. B. H., & Kyzas, G. Z. (2020). Petroleum Hydrocarbon Removal from Wastewaters: A Review. Processes, 8(4), 447. https://doi.org/10.3390/pr8040447

Mohammadi, T., Kazemimoghadam, M., & Saadabadi, M. (2003). Modeling of membrane fouling and flux decline in reverse osmosis during separation of oil in water emulsions. Desalination, 157(1–3), 369–375. https://doi.org/10.1016/S0011-9164(03)00419-3

Mondal, S., & Wickramasinghe, S. R. (2008). Produced water treatment by nanofiltration and reverse osmosis membranes. Journal of Membrane Science, 322(1), 162–170. https://doi.org/10.1016/j.memsci.2008.05.039

Morales-Caselles, C., Yunker, M. B., & Ross, P. S. (2017). Identification of Spilled Oil from the MV Marathassa (Vancouver, Canada 2015) Using Alkyl PAH Isomer Ratios. Archives of Environmental Contamination and Toxicology, 73(1), 118–130. https://doi.org/10.1007/s00244-017-0390-0

Mutamim, N. S. A., Noor, Z. Z., Hassan, M. A. A., & Olsson, G. (2012). Application of membrane bioreactor technology in treating high strength industrial wastewater: A performance review. Desalination, 305, 1–11. https://doi.org/10.1016/j.desal.2012.07.033

Naim, R., Pei Sean, G., Nasir, Z., Mokhtar, N. M., & Safiah Muhammad, N. A. (2021). Recent Progress and Challenges in Hollow Fiber Membranes for Wastewater Treatment and Resource Recovery. Membranes, 11(11), 839. https://doi.org/10.3390/membranes11110839

National Center for Biotechnology Information. (2022a). PubChem Compound Summary for CID 6734, Acenaphthene. https://pubchem.ncbi.nlm.nih.gov/compound/Acenaphthene#section=Information-Sources

National Center for Biotechnology Information. (2022b). PubChem Compound Summary for CID 6853, Fluorene. https://pubchem.ncbi.nlm.nih.gov/compound/Fluorene#section=Entrez-Crosslinks

National Center for Biotechnology Information. (2022c). PubChem Compound Summary for CID 7002, 1-Methylnaphthalene. https://pubchem.ncbi.nlm.nih.gov/compound/1-methylnaphthalene#section=Solubility

National Center for Biotechnology Information. (2022d). PubChem Compound Summary for CID 9154, Fluoranthene. https://pubchem.ncbi.nlm.nih.gov/compound/Fluoranthene

National Center for Biotechnology Information. (2022e). PubChem Compound Summary for CID 9171, Chrysene. https://pubchem.ncbi.nlm.nih.gov/compound/Chrysene#section=Solubility

National Center for Biotechnology Information. (2022f). PubChem Compound Summary for CID 31423, Pyrene. https://pubchem.ncbi.nlm.nih.gov/compound/Pyrene#section=Solubility

Nerubenko, G. (2015). Underwater Technique for Prevention of Oil Pollution. Journal of Water Resources and Ocean Science, 4, 1. https://doi.org/10.11648/j.wros.s.2015040101.11

Norouzbahari, S., Roostaazad, R., & Hesampour, M. (2009). Crude oil desalter effluent treatment by a hybrid UF/RO membrane separation process. Desalination, 238(1), 174–182. https://doi.org/10.1016/j.desal.2008.01.045

Nyström, M. (1991). Ultrafiltration of O/W emulsions stabilized by limiting amounts of tall oil. Colloids and Surfaces, 57(1), 99–114. https://doi.org/10.1016/0166-6622(91)80183-O

Obotey Ezugbe, E., & Rathilal, S. (2020). Membrane Technologies in Wastewater Treatment: A Review. Membranes, 10(5), 89. https://doi.org/10.3390/membranes10050089

OSPAR commission. (2015). Assessment of the discharges, spills and emissions from offshore oil and gas operations on the United Kingdom Continental Shelf 2009-2013. Available online: https://www.ospar.org/documents?v=33827. (accessed on 10 September 2022)

Otitoju, T. A., Ahmad, A. L., & Ooi, B. S. (2017). Polyethersulfone composite hollow-fiber membrane prepared by in-situ growth of silica with highly improved oily wastewater separation performance. Journal of Polymer Research, 24(8), 123. https://doi.org/10.1007/s10965-017-1268-6

Padaki, M., Surya Murali, R., Abdullah, M. S., Misdan, N., Moslehyani, A., Kassim, M. A., Hilal, N., & Ismail, A. F. (2015). Membrane technology enhancement in oil–water separation. A review. Desalination, 357, 197–207. https://doi.org/10.1016/j.desal.2014.11.023

Park, S., Yeon, K.-M., Moon, S., & Kim, J.O. (2018). Enhancement of operating flux in a membrane bio-reactor coupled with a mechanical sieve unit. Chemosphere, 191, 573–579. https://doi.org/10.1016/j.chemosphere.2017.10.079

Pendashteh, A. R., Fakhru'l-Razi, A., Madaeni, S. S., Abdullah, L. C., Abidin, Z. Z., & Biak, D. R. A. (2011). Membrane foulants characterization in a membrane bioreactor (MBR) treating hypersaline oily wastewater. Chemical Engineering Journal, 168(1), 140–150. https://doi.org/10.1016/j.cej.2010.12.053

Penela-Arenaz, M., Bellas, J., & Vázquez, E. (2009). Chapter Five: Effects of the Prestige Oil Spill on the Biota of NW Spain: 5 Years of Learning. In Advances in Marine Biology (Vol. 56, pp. 365–396). Academic Press. https://doi.org/10.1016/S0065-2881(09)56005-1

Peng, H., & Tremblay, A. Y. (2008). Membrane regeneration and filtration modeling in treating oily wastewaters. Journal of Membrane Science, 324(1), 59–66. https://doi.org/10.1016/j.memsci.2008.06.062

Perhar, G., & Arhonditsis, G. B. (2014). Aquatic ecosystem dynamics following petroleum hydrocarbon perturbations: A review of the current state of knowledge. Journal of Great Lakes Research, 40, 56-72. https://doi.org/10.1016/j.jglr.2014.05.013

Polak, J., & Lu, B. C.Y. (1973). Mutual Solubilities of Hydrocarbons and Water at 0 and 25 °C. Canadian Journal of Chemistry, 51(24), 4018–4023. https://doi.org/10.1139/v73-599

Porcelli, N., & Judd, S. (2010). Chemical cleaning of potable water membranes: A review. Separation and Purification Technology, 71(2), 137–143. https://doi.org/10.1016/j.seppur.2009.12.007

Potts, D. E., Ahlert, R. C., & Wang, S. S. (1981). A critical review of fouling of reverse osmosis membranes. Desalination, 36(3), 235–264. https://doi.org/10.1016/S0011-9164(00)88642-7

Psoch, C., & Schiewer, S. (2006). Resistance analysis for enhanced wastewater membrane filtration. Journal of Membrane Science, 280(1–2), 284–297. https://doi.org/10.1016/j.memsci.2006.01.030

Radjenovic, J., Matošić, M., Mijatović, I., Petrovic, M., & Barcelo, D. (2008). Membrane Bioreactor (MBR) as an Advanced Wastewater Treatment Technology. Handbook of Environmental Chemistry, Volume 5: Water Pollution, 5. https://doi.org/10.1007/698_5_093

Ravazzini, A. M., van Nieuwenhuijzen, A. F., & van der Graaf, J. H. M. J. (2005). Direct ultrafiltration of municipal wastewater: Comparison between filtration of raw sewage and primary clarifier effluent. Desalination, 178(1–3), 51–62. https://doi.org/10.1016/j.desal.2004.11.028

Razavi, S. M. R., & Miri, T. (2015). A real petroleum refinery wastewater treatment using hollow fiber membrane bioreactor (HF-MBR). Journal of Water Process Engineering, 8, 136–141. https://doi.org/10.1016/j.jwpe.2015.09.011

Reyhani, A., Rekabdar, F., Hemmati, M., SafeKordi, A. A., & Ahmadi, M. (2013). Optimization of conditions in ultrafiltration treatment of produced water by polymeric membrane using Taguchi approach. Desalination and Water Treatment, 51(40–42), 7499–7508. https://doi.org/10.1080/19443994.2013.776505

Rezvanpour, A., Roostaazad, R., Hesampour, M., Nyström, M., & Ghotbi, C. (2009). Effective factors in the treatment of kerosene–water emulsion by using UF membranes. Journal of Hazardous Materials, 161(2–3), 1216–1224. https://doi.org/10.1016/j.jhazmat.2008.04.074

Romero, C. A., & Davis, R. H. (1991). Experimental verification of the shear-induced hydrodynamic diffusion model of crossflow microfiltration. Journal of Membrane Science, 62(3), 249–273. https://doi.org/10.1016/0376-7388(91)80042-5

Rutter, A., Zeeb, B., & Matovic, D. (2014). Sustainable remediation of petroleum hydrocarbons using phytotechnologies. 66. https://www.eralberta.ca/wp-content/uploads/2017/06/CCEMC-Sustainable-Remediation-Final-Report.pdf

Sadeghi, F., Mehrnia, M. R., Nabizadeh, R., Bahadori, M. B., & Sarrafzadeh, M. H. (2017). MBR technology: A practical approach for petrochemical wastewater treatment. Petroleum Science and Technology, 35(3), 222–228. https://doi.org/10.1080/10916466.2011.572108

Safazadeh Haghighi, L. (2011). Determination of Fouling Mechanisms for Ultrafiltration of Oily Wastewater, Master's thesis, University of Cincinnati. https://etd.ohiolink.edu/apexprod/rws_olink/r/1501/10?clear=10&p10_accession_num=ucin1321371722

Saini, B., Sinha, M. K., & Dash, S. K. (2019). Mitigation of HA, BSA and oil/water emulsion fouling of PVDF Ultrafiltration Membranes by SiO_2-g-PEGMA nanoparticles. Journal of Water Process Engineering, 30, 100603. https://doi.org/10.1016/j.jwpe.2018.03.018

Salahi, A., Gheshlaghi, A., Mohammadi, T., & Madaeni, S. S. (2010). Experimental performance evaluation of polymeric membranes for treatment of an industrial oily wastewater. Desalination, 262(1–3), 235–242. https://doi.org/10.1016/j.desal.2010.06.021

Salahi, A., Badrnezhad, R., Abbasi, M., Mohammadi, T., & Rekabdar, F. (2011). Oily wastewater treatment using a hybrid UF/RO system. Desalination and Water Treatment, 28(1–3), 75–82. https://doi.org/10.5004/dwt.2011.2204

Salahi, A., Mohammadi, T., Behbahani, R. M., & Hemmati, M. (2015). Experimental investigation and modeling of industrial oily wastewater treatment using modified polyethersulfone ultrafiltration hollow fiber membranes. Korean Journal of Chemical Engineering, 32(6), 1101–1118. https://doi.org/10.1007/s11814-014-0310-1

Sanghamitra, P., Mazumder, D., & Mukherjee, S. (2021). Treatment of wastewater containing oil and grease by biological method- a review. Journal of Environmental Science and Health, Part A, 56(4), 394–412. https://doi.org/10.1080/10934529.2021.1884468

Schwinge, J., Neal, P. R., Wiley, D. E., Fletcher, D. F., & Fane, A. G. (2004). Spiral wound modules and spacers. Journal of Membrane Science, 242(1–2), 129–153. https://doi.org/10.1016/j.memsci.2003.09.031

Sempere, C. (2015). Nanofluidic insight into energy harvesting and desalination, PhD thesis, Université Claude Bernard - Lyon I. https://doi.org/10/document

Sentana, I., De La Rubia, M. A., Rodríguez, M., Sentana, E., & Prats, D. (2009). Removal of natural organic matter by cationic and anionic polyacrylonitrile membranes. The effect of pressure, ionic strength and pH. Separation and Purification Technology, 68(3), 305–311. https://doi.org/10.1016/j.seppur.2009.05.017

Seyed Shahabadi, S. M., & Reyhani, A. (2014). Optimization of operating conditions in ultrafiltration process for produced water treatment via the full factorial design methodology. Separation and Purification Technology, 132, 50–61. https://doi.org/10.1016/j.seppur.2014.04.051

Sharghi, E. A., Shourgashti, A., & Bonakdarpour, B. (2020). Considering a membrane bioreactor for the treatment of vegetable oil refinery wastewaters at industrially relevant organic loading rates. Bioprocess and Biosystems Engineering, 43(6), 981–995. https://doi.org/10.1007/s00449-020-02294-9

Shi, C., Yan, B., Xie, L., Zhang, L., Wang, J., Takahara, A., & Zeng, H. (2016). Long-Range Hydrophilic Attraction between Water and Polyelectrolyte Surfaces in Oil. Angewandte Chemie International Edition, 55(48), 15017–15021. https://doi.org/10.1002/anie.201608219

Shi, X., Lefebvre, O., Ng, K. K., & Ng, H. Y. (2014). Sequential anaerobic–aerobic treatment of pharmaceutical wastewater with high salinity. Bioresource Technology, 153, 79–86. https://doi.org/10.1016/j.biortech.2013.11.045

Singh, V., Purkait, M. K., & Das, C. (2011). Cross-Flow Microfiltration of Industrial Oily Wastewater: Experimental and Theoretical Consideration. Separation Science and Technology, 46(8), 1213–1223. https://doi.org/10.1080/01496395.2011.560917

SL Ross. (2005). Transfer of decanting technology research to oil spill response organizations and regulators. 63. Available online: https://www.bsee.gov/sites/bsee.gov/files/osrr-oil-spill-response-research/512aa.pdf (accessed on 18 January 2022)

Soltani, S., Mowla, D., Vossoughi, M., & Hesampour, M. (2010). Experimental investigation of oily water treatment by membrane bioreactor. Desalination, 250(2), 598–600. https://doi.org/10.1016/j.desal.2009.09.031

Song, H., Yu, H., Zhu, L., Xue, L., Wu, D., & Chen, H. (2017). Durable hydrophilic surface modification for PTFE hollow fiber membranes. Reactive and Functional Polymers, 114, 110–117. https://doi.org/10.1016/j.reactfunctpolym.2017.03.010

Song, X., Chen, B., Liu, B., Lye, L. M., Ye, X., Nyantekyi-Kwakye, B., & Zhang, B. (2022). Impacts of Frazil Ice on the Effectiveness of Oil Dispersion and Migration of Dispersed Oil | Environmental Science & Technology. https://doi.org/10.1021/acs.est.1c04014

Souza, E. C., Vessoni-Penna, T. C., & de Souza Oliveira, R. P. (2014). Biosurfactant-enhanced hydrocarbon bioremediation: An overview. International Biodeterioration & Biodegradation, 89, 88–94. https://doi.org/10.1016/j.ibiod.2014.01.007

Souza, J. S. B., Júnior, J. F., Simonelli, G., Souza, J. R., Góis, L. M. N., & Santos, L. C. L. (2020). Removal of oil contents and salinity from produced water using microemulsion. Journal of Water Process Engineering, 38, 101548. https://doi.org/10.1016/j.jwpe.2020.101548

Soydemir, G., Gurol, M. D., Hocaoğlu, S. M., & Karagündüz, A. (2020). Fouling mechanisms of membrane filtration of mixed microalgal biomass grown in wastewater. Water Science and Technology, 81(10), 2127–2139. https://doi.org/10.2166/wst.2020.268

Spies, R. B., Mukhtasor, M., & Burns, K. A. (2017). The Montara Oil Spill: A 2009 Well Blowout in the Timor Sea. Archives of Environmental Contamination and Toxicology, 73(1), 55–62. https://doi.org/10.1007/s00244-016-0356-7

Stephenson, T., Brindle, K., Judd, S., & Jefferson, B. (2000). Membrane Bioreactors for Wastewater Treatment. IWA Publishing.

Sterlitech Corporation. (2017). Crossflow Filtration Handbook. https://cdn.thomasnet.com/ccp/10096818/249376.pdf

Sun, C., Leiknes, T., Weitzenböck, J., & Thorstensen, B. (2010). Salinity effect on a biofilm-MBR process for shipboard wastewater treatment. Separation and Purification Technology, 72(3), 380–387. https://doi.org/10.1016/j.seppur.2010.03.010

Syafiuddin, A., & Boopathy, R. (2021). A review of polycyclic aromatic hydrocarbons and their substitutions in full-scale wastewater treatment plants. Environmental Quality Management, 31(1), 21–37. https://doi.org/10.1002/tqem.21694

Takadono, S., Iwahori, H., Yabushita, T., Imamura, Y., Ishizuka, H., & Tamaru, S. (1984). Treatment of highly fouling waste waters with tubular membrane systems. Desalination, 49(3), 347–355. https://doi.org/10.1016/0011-9164(84)85042-0

Tanudjaja, H. J., Hejase, C. A., Tarabara, V. V., Fane, A. G., & Chew, J. W. (2019). Membrane-based separation for oily wastewater: A practical perspective. Water Research, 156, 347–365. https://doi.org/10.1016/j.watres.2019.03.021

Tran, V. H., Lim, S., Choi, P. J., An, A. K., Han, D. S., Phuntsho, S., & Shon, H. (2021). Submerged versus side-stream osmotic membrane bioreactors using an outer-selective hollow fiber osmotic membrane for desalination. Desalination, 515, 115196. https://doi.org/10.1016/j.desal.2021.115196

Tummons, E. N., Chew, J. W., Fane, A. G., & Tarabara, V. (2017). Ultrafiltration of saline oil-in-water emulsions stabilized by an anionic surfactant_ Effect of surfactant concentration and divalent counterions. Journal of Membrane Science, 12. https://doi.org/10.1016/j.memsci.2017.05.012

Tummons, E., Han, Q., Tanudjaja, H. J., Hejase, C. A., Chew, J. W., & Tarabara, V. V. (2020). Membrane fouling by emulsified oil: A review. Separation and Purification Technology, 248, 116919. https://doi.org/10.1016/j.seppur.2020.116919

Ueda, T., Hata, K., Kikuoka, Y., & Seino, O. (1997). Effects of aeration on suction pressure in a submerged membrane bioreactor. Water Research (Oxford), 31(3), 489–494. https://doi.org/10.1016/S0043-1354(96)00292-8

Ullah, A., Starov, V. M., Naeem, M., & Holdich, R. G. (2011). Microfiltration of deforming oil droplets on a slotted pore membrane and sustainable flux rates. Journal of Membrane Science, 382(1), 271–277. https://doi.org/10.1016/j.memsci.2011.08.017

Ullah, A., Tanudjaja, H. J., Ouda, M., Hasan, S. W., & Chew, J. W. (2021). Membrane fouling mitigation techniques for oily wastewater: A short review. Journal of Water Process Engineering, 43, 102293. https://doi.org/10.1016/j.jwpe.2021.102293

USEPA. (1999). Understanding Oil Spills and Oil Spill Response. Available online: https://www.epa.gov/sites/default/files/2018-01/documents/ospguide99.pdf (accessed on 5 May 2022)

USEPA. (2016). Oil and gas extraction effluent guidelines. Available online: https://www.epa.gov/eg/oil-and-gas-extraction-effluent-guidelines (accessed on 10 September 2022)

Varjani, S., Joshi, R., Srivastava, V. K., Ngo, H. H., & Guo, W. (2020). Treatment of wastewater from petroleum industry: Current practices and perspectives. Environmental Science and Pollution Research, 27(22), 27172–27180. https://doi.org/10.1007/s11356-019-04725-x

Veil, J. (2015). U.S. Produced Water Volumes and Management Practices in 2012. Available online: http://www.veilenvironmental.com/publications/pw/final_report_CO_note.pdf (accessed on 10 July 2021)

Vinther, F., Pinelo, M., Brøns, M., Jonsson, G., & Meyer, A. S. (2014). Mathematical modelling of dextran filtration through hollow fibre membranes. Separation and Purification Technology, 125, 21-36. http://dx.doi.org/10.1016/j.seppur.2014.01.034

Visvikis, I. D., & Panayides, P. M. (Eds.). (2017). Shipping Operations Management (Vol. 4). Springer International Publishing. https://doi.org/10.1007/978-3-319-62365-8

Wan, C. F., Yang, T., Lipscomb, G. G., Stookey, D. J., & Chung, T.S. (2017). Design and fabrication of hollow fiber membrane modules. Journal of Membrane Science, 538, 96–107. https://doi.org/10.1016/j.memsci.2017.05.047

Wang, W. (2008). Toxicity Assessment of PAHs and Metals to Bacteria and the Roles of Soil Bacteria in Phytoremediation of Petroleum Hydrocarbons. https://uwspace.uwaterloo.ca/handle/10012/4068

Wang, Y., Chen, X., Zhang, J., Yin, J., & Wang, H. (2009). Investigation of microfiltration for treatment of emulsified oily wastewater from the processing of petroleum products. 3(249), 1223–1227. https://doi.org/10.1016/j.desal.2009.06.033

Wei, X., Zhang, S., Han, Y., & Wolfe, F. A. (2019). Treatment of petrochemical wastewater and produced water from oil and gas. Water Environment Research, 91(10), 1025–1033. https://doi.org/10.1002/wer.1172

Wu, J., Le-Clech, P., Stuetz, R. M., Fane, Anthony. G., & Chen, V. (2008). Effects of relaxation and backwashing conditions on fouling in membrane bioreactor. Journal of Membrane Science, 324(1), 26–32. https://doi.org/10.1016/j.memsci.2008.06.057

Wu, P., Jiang, L. Y., He, Z., & Song, Y. (2017). Treatment of metallurgical industry wastewater for organic contaminant removal in China: Status, challenges, and perspectives. Environmental Science: Water Research & Technology, 3(6), 1015–1031. https://doi.org/10.1039/C7EW00097A

Xu, Q., Yang, Y., Wang, X., Wang, Z., Jin, W., Huang, J., & Wang, Y. (2012). Atomic layer deposition of alumina on porous polytetrafluoroethylene membranes for enhanced hydrophilicity and separation performances. Journal of Membrane Science, 415–416, 435–443. https://doi.org/10.1016/j.memsci.2012.05.031

Yang, C. (2007). Electrochemical coagulation for oily water demulsification. Separation and Purification Technology, 54(3), 388–395. https://doi.org/10.1016/j.seppur.2006.10.019

Yang, Y., Raza, A., Banat, F., & Wang, K. (2018). The separation of oil in water (O/W) emulsions using polyether sulfone & nitrocellulose microfiltration membranes. Journal of Water Process Engineering, 25, 113–117. https://doi.org/10.1016/j.jwpe.2018.07.007

Yang, Z., Shah, K., Laforest, S., Hollebone, B. P., Situ, J., Crevier, C., Lambert, P., Brown, C. E., & Yang, C. (2020). Occurrence and weathering of petroleum hydrocarbons deposited on the shoreline of the North Saskatchewan River from the 2016 Husky oil spill. Environmental Pollution, 258, 113769. https://doi.org/10.1016/j.envpol.2019.113769

Ye, Y., Sim, L. N., Herulah, B., Chen, V., & Fane, A. G. (2010). Effects of operating conditions on submerged hollow fibre membrane systems used as pre-treatment for seawater reverse osmosis. Journal of Membrane Science, 365(1), 78–88. https://doi.org/10.1016/j.memsci.2010.08.038

Yim, U. H., Khim, J. S., Kim, M., Jung, J.-H., & Shim, W. J. (2017). Environmental Impacts and Recovery After the Hebei Spirit Oil Spill in Korea. Archives of Environmental Contamination and Toxicology, 73(1), 47–54. https://doi.org/10.1007/s00244-017-0375-z

Young, J., Skewes, T., Lyne, V., Hook, S. E., Revill, A., Condie, S., Newman, S., Wakefield, C., & Molony, B. (2011). A Review of the Fisheries Potentially Affected by the Montara Oil Spill off Northwest Australia and Potential Toxicological Effects. Report to PTTEP Australasia.

Yu, L., Han, M., & He, F. (2017). A review of treating oily wastewater. Arabian Journal of Chemistry, 10, S1913–S1922. https://doi.org/10.1016/j.arabjc.2013.07.020

Yuliwati, E., Ismail, A. F., Lau, W. J., Ng, B. C., Mataram, A., & Kassim, M. A. (2012). Effects of process conditions in submerged ultrafiltration for refinery wastewater treatment: Optimization of operating process by response surface methodology. Desalination, 287, 350–361. https://doi.org/10.1016/j.desal.2011.08.051

Yusuf, Z., Abdul Wahab, N., & Sahlan, S. (2016). Fouling control strategy for submerged membrane bioreactor filtration processes using aeration airflow, backwash, and relaxation: A review. Desalination and Water Treatment, 57(38), 17683–17695. https://doi.org/10.1080/19443994.2015.1086893

Zare, M., Zokaee Ashtiani, F., & Fouladitajar, A. (2013). CFD modeling and simulation of concentration polarization in microfiltration of oil–water emulsions; Application of an Eulerian multiphase model. Desalination, 324, 37–47. https://doi.org/10.1016/j.desal.2013.05.022

Zhang, J., Chua, H. C., Zhou, J., & Fane, A. G. (2006). Factors affecting the membrane performance in submerged membrane bioreactors. Journal of Membrane Science, 284(1–2), 54–66. https://doi.org/10.1016/j.memsci.2006.06.022

Zheng, J., Liu, B., Ping, J., Chen, B., Wu, H., & Zhang, B. (2015). Vortex- and Shaker-Assisted Liquid–Liquid Microextraction (VSA-LLME) Coupled with Gas Chromatography and Mass Spectrometry (GC-MS) for Analysis of 16 Polycyclic Aromatic Hydrocarbons (PAHs) in Offshore Produced Water. Water, Air, & Soil Pollution, 226(9), 318. https://doi.org/10.1007/s11270-015-2575-3

Zhidong, L., Na, L., Honglin, Z., & Dan, L. (2009). Study of an A/O Submerged Membrane Bioreactor for Oil Refinery Wastewater Treatment. Petroleum Science and Technology, 27(12), 1274–1285. https://doi.org/10.1080/10916460802455228

Zhu, X., Tu, W., Wee, K.-H., & Bai, R. (2014). Effective and low fouling oil/water separation by a novel hollow fiber membrane with both hydrophilic and oleophobic surface properties. Journal of Membrane Science, 466, 36–44. https://doi.org/10.1016/j.memsci.2014.04.038

Zsirai, T., Buzatu, P., Aerts, P., & Judd, S. (2012). Efficacy of relaxation, backflushing, chemical cleaning and clogging removal for an immersed hollow fibre membrane bioreactor. Water Research, 46(14), 4499–4507. https://doi.org/10.1016/j.watres.2012.05.004

APPENDIX: Samples Taken from the Experiments

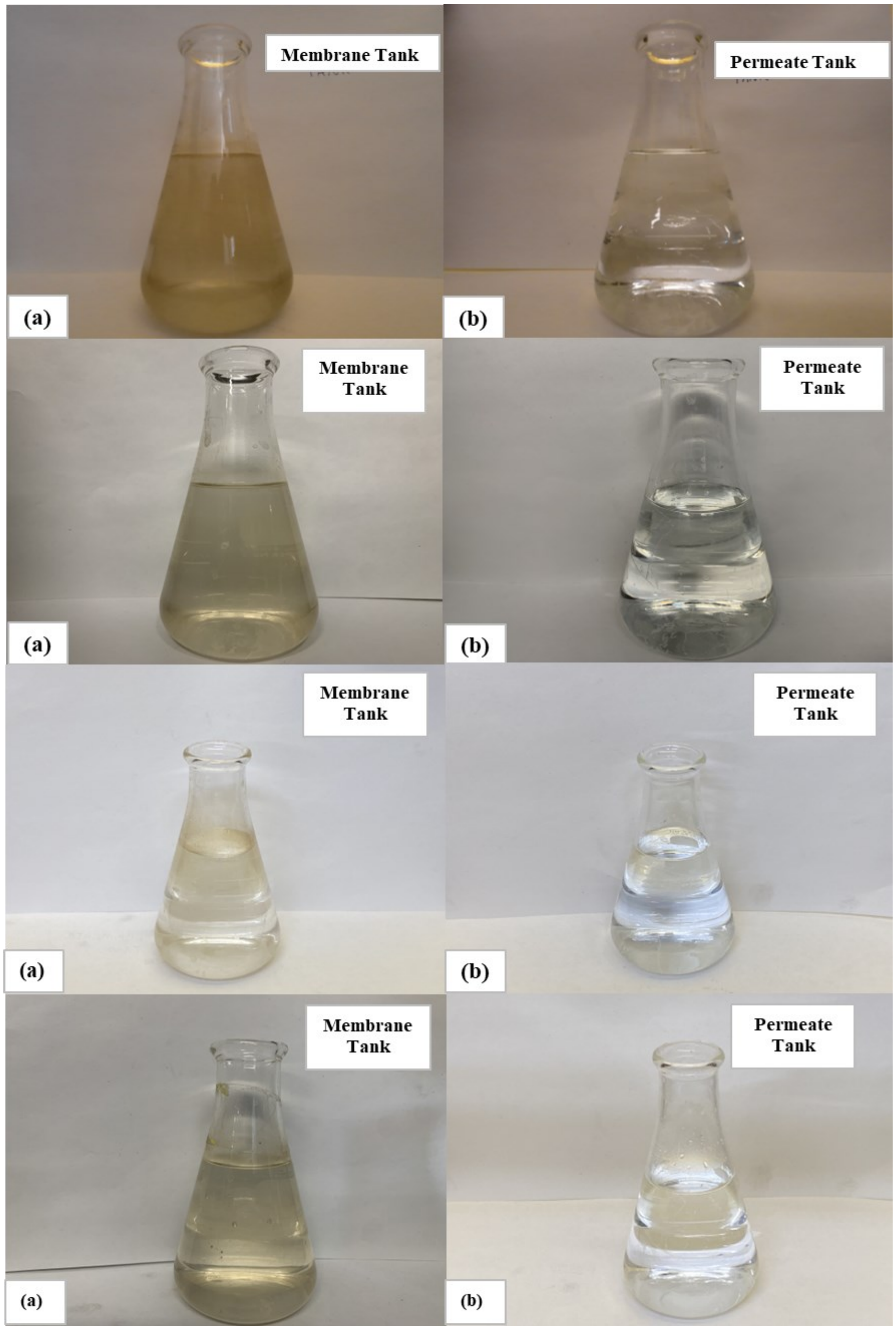

Figure A1 Oily wastewater containing 50 ppm CLD (a) membrane tank (b) permeate tank

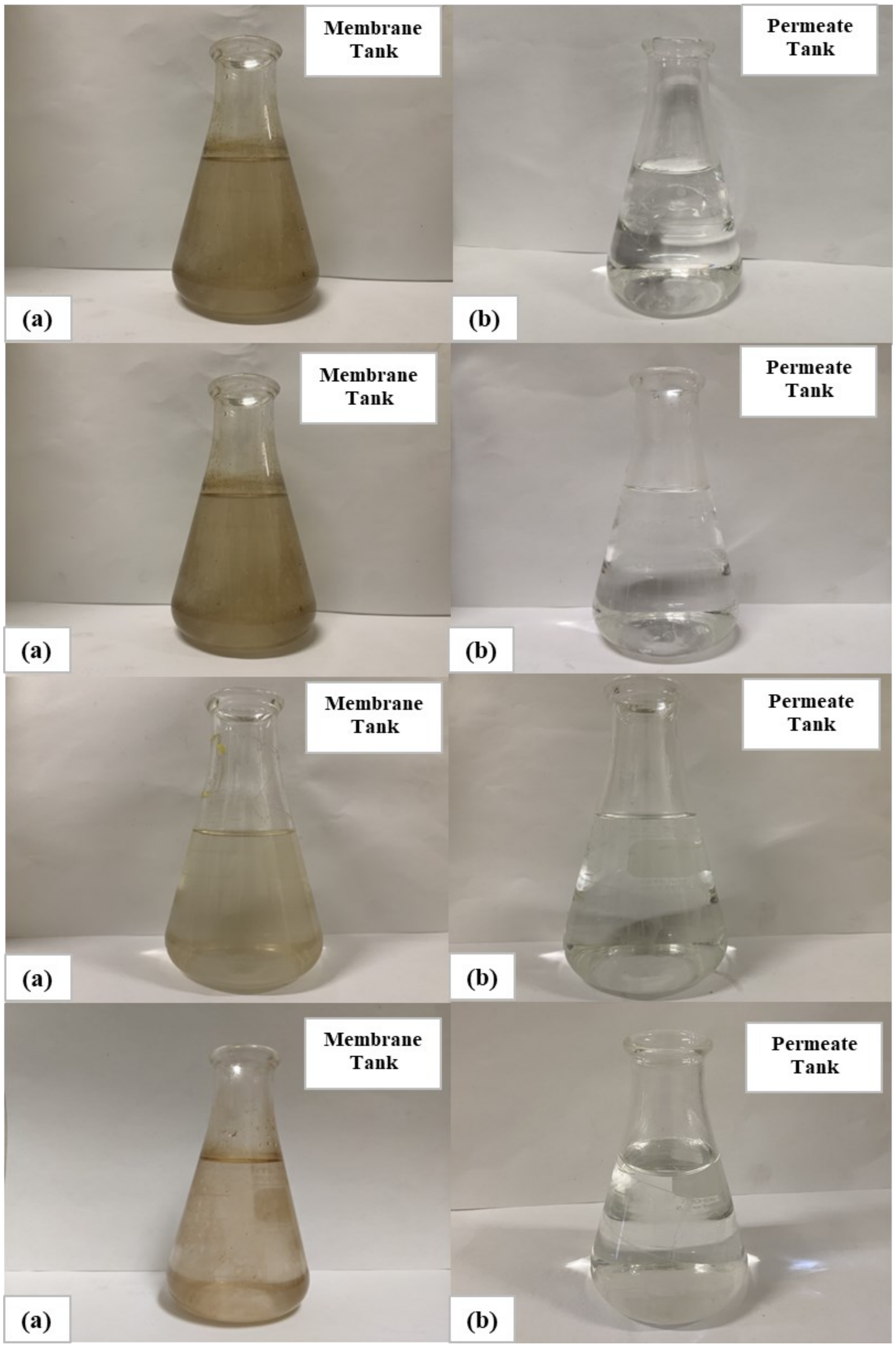

Figure A2 Oily wastewater containing 100 ppm CLD (a) membrane tank (b) permeate tank

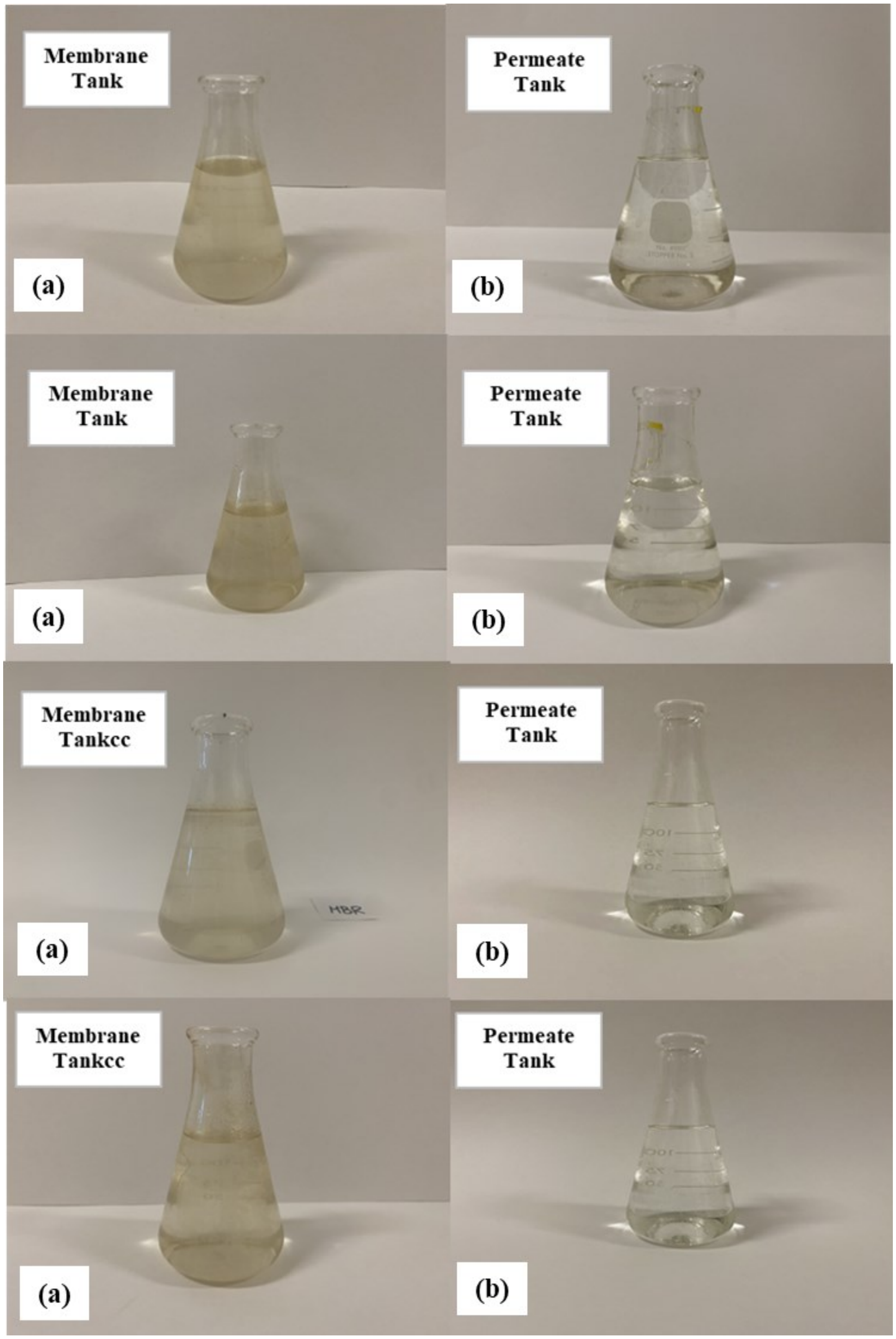

Figure A3 Oily wastewater containing 50 ppm VLSFO (a) membrane tank (b) permeate tank

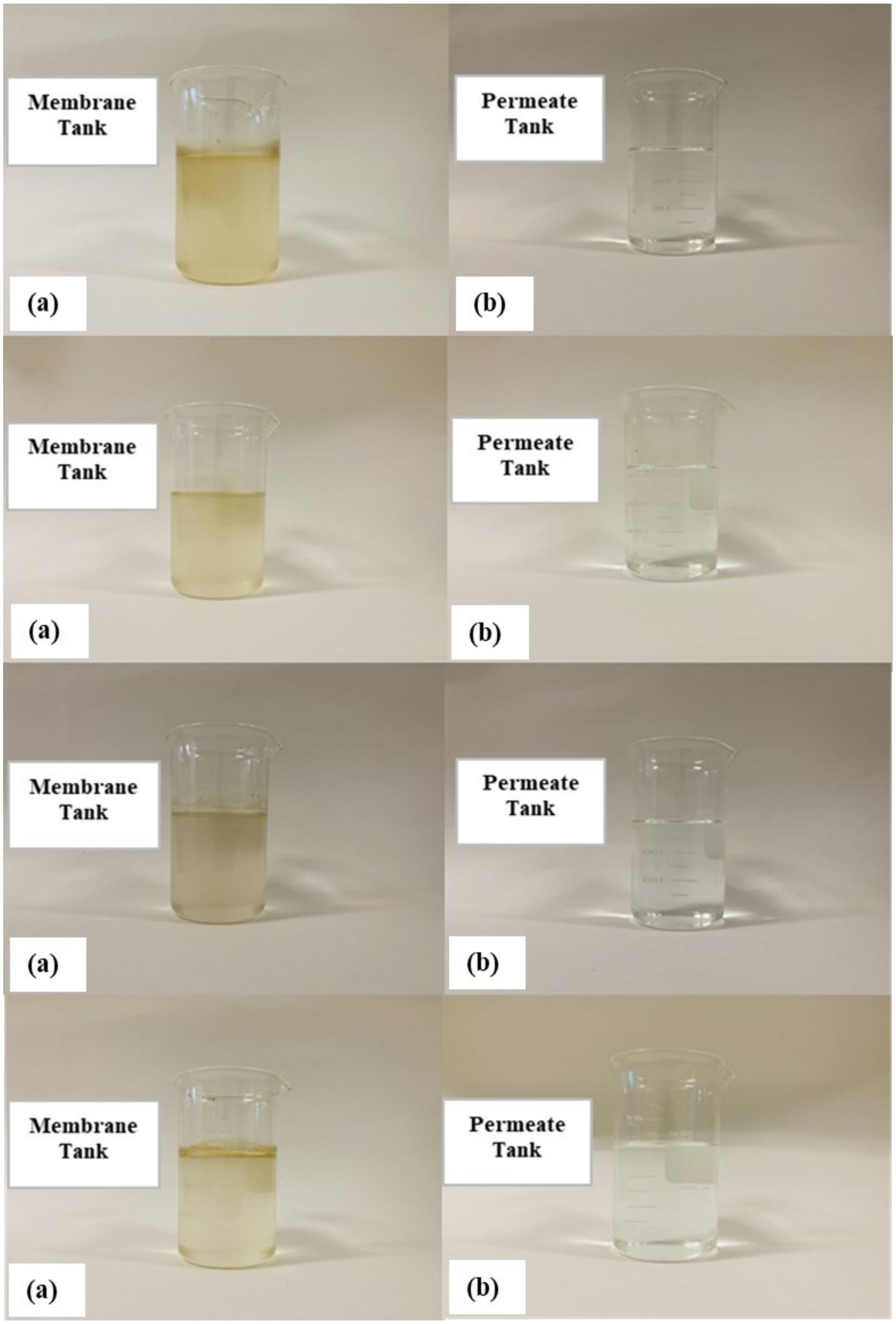

Figure A4 Oily wastewater containing 100 ppm VLSFO (a) membrane tank (b) permeate tank