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# Introductory Chapter: Osmotically Driven Membrane Processes

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Felecia Nave, Raghava Kommalapati and  
Audie Thompson

Additional information is available at the end of the chapter

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## 1. Forward osmosis fundamentals

Global climate patterns and urban growth are two of the many factors that have affected the world's water resources. During the twentieth century, the population of the world tripled, and it is predicted to increase by another 15–20% in the next 50 years [1, 2]. The demand for fresh potable water correlates with the increase in the world's population, thus access to safe and sufficient drinking water is now an international aim. Sadly, over 1 billion people across the world currently have limited to no access to drinking water [3]. In particular, the demand for water drastically outweighs the availability of water in some Middle Eastern countries and even within the United States, in states such as California that has recently experienced droughts [4]. Further, urbanization throughout the world has also impacted groundwater resources [5], and this controversy has led to surging interest in the efficiency and practicality of ocean water desalination [6].

Desalination is the process of obtaining drinking water by removing salt ions, minerals, and other undesired contaminants from seawater [7], and currently, there is an increasing interest in using FO in desalination. In arid regions of the world, such as the Mediterranean and the Middle East, desalination research has made great strides over the past 30 years [8]. In fact, there are approximately 14,000 desalination plants in 150 countries with a production of millions of gallons per day [8]. In countries, such as Saudi Arabia and the United Arab Emirates, 70% of water supplies are dependent on desalination. Hence, energy production is concurrently linked to the production of freshwater, as desalination of seawater requires more energy than transportation of water from a lake or river [9]. It is also important to note that nuclear plants and other energy sources (coal or oil) require 20–50 K gallons of water per megawatt-hour of electricity produced [10]. Furthermore, gasoline vehicles, plug-in vehicles, ethanol-running vehicles and hydrogen-fuel cell vehicles all consume gallons of water to operate. Thus, the demand for water is intrinsically tied to energy and sustainable practices

and processes must be used. Discovering energetically efficient methods to produce and reuse water is pertinent in providing strategies to combat the energy consumption demands. Additionally, industrial plants consume a drastic amount of water for their industrial processes, and 70% of fresh water is utilized in agricultural processes [11]. Therefore, water shortages will hinder many areas of human daily activity and existence.

Most water-related technologies are based on advanced materials, advanced manufacturing technologies, biotechnology, and integrated filtration systems. Therefore, research and development of new materials with tailored properties and nanomaterials are necessary to meet the water demands and provide connections between eco-efficiency, performance, processing, recyclability, costs, and water reuse. Although the development of membrane technology for producing clean water in wastewater treatment and desalination is vital, there are challenges that must be further addressed in all water filtration processes [12, 13]. Water-selective membranes have gained vast interest for their advantages like high energy efficiency, reasonable cost, and environmental sustainability. The ideal water-selective membranes are fabricated to have high water permeability, selectivity, as well as stability [14]. However, major constraints include operational fouling, waste residue disposal, cost, and acceptance by utility organizations and the public.

The current and most widely used water purification is reverse osmosis (RO)—a membrane-based separation process that removes salts, microbial constituents, both organic and inorganic compounds from water and has been used extensively in a variety of fields including desalination of seawater, ultrapure water production, and wastewater treatment [15, 16]. RO goes against the laws of nature and uses pressure to force a solvent through the membrane, which retains the solute on one side and allows the pure solvent to pass to the other side. Since its discovery, RO has become a very useful process when it comes to removing salt ions from a solution.

There has been an increased focus on membrane technology research because of the high efficiency and low-cost solutions for water purification. Currently, forward osmosis (FO) systems are seen as favorable alternatives to RO systems, as they have been also utilized in electricity generation, food processing [11], industrial wastewater, and add produced water treatment [17–19]. In nature, when two solutions are separated by a semipermeable membrane, the solvent molecules will tend to move through the membrane into the region of higher solute concentration until equilibrium is reached. FO separates two solutions with different concentrations using the natural osmotic pressure difference. The osmotic gradient is the driving force instead of externally applied pressure.

Even though RO systems have dominated the water purification arena for decades, FO systems offer an advantage of rejecting a wide range of contaminants. FO systems experience less fouling than RO systems; therefore, a membrane with anti-fouling properties could be efficient and beneficial. Within the RO process, the saline water, which has a high salt concentration, is forced through a membrane to a region of low solute concentrate by applying pressure in excess of osmotic pressure [20, 21], where the osmotic pressure is the minimum pressure needed to prevent the water molecules from moving back to the feed side from the permeate side. This occurs when the hydrostatic pressure differential resulting from the concentration

changes on both sides of the semipermeable membrane is equal to the osmotic pressure of the solute [21]. The semipermeable membrane allows the passage of water but not salt ions. The feed water must pass through a very narrow passage as a result of the way the membrane is packaged. This causes for an initial treatment phase, where fine particulates or suspended solids must be removed to prevent fouling. In contrast, the FO system will have higher productivity and be considered an energy saving device since no external pressure is required. However, a major and unresolved challenge in FO remains an efficient draw solution that could result in high flux and reconstituted using a low-energy separation process which will be discussed later.

Two key factors in FO utilization are selecting the membrane and appropriate draw solute (DS). The DS should be non-toxic, generate high osmotic pressure, and be easily regenerated [22]. Continuous reconcentration is required to sustain the FO driving force to purify water. NaCl, MgCl<sub>2</sub>, CaCl<sub>2</sub>, and MgSO<sub>4</sub> are commonly used DSs; however, they are energy intensive and consequently costly [22, 23]. Alternatively, the DS can be treated wastewater effluent brine or seawater; the diluted DS will lower the energy demand [22]. Other limitations are the diffusion of the DS into the feed solution, low water flux compared to RO, membrane fouling, and concentration polarization. Therefore, many researchers are investigating alternative DSs.

### 1.1. Wastewater and water recycling

Wastewater sources include municipal and industrial plants and consume a drastic amount of water for their industrial processes. Some plants also produce oily wastewater end products. The industries that account for oil in water emulsions are petroleum, pharmaceutical, polymer, leather, polish, cosmetic, food, polymer, textile, agriculture, prints, and paper [24]. Helen Wake reports that oil refineries in European and Middle Eastern countries alone produce over 2 billion tons of wastewater [25]. This strikes as a major ecological problem, due to the discharge of oily wastewater into the ecosystem [25]. Furthermore, a principal fraction of oil/water emulsions' treatment technologies is often ineffective and expensive [24].

Produced water (PW) is generated during oil and gas production and is the biggest waste stream in the energy industries [26, 27]. Therefore, PW is contaminated with oils and salts of organic and inorganic compounds [27]. Releasing PW onto nature has an environmental impact and is a noteworthy issue of ecological concern. Ordinarily, PW is treated through various physical, chemical, and biological strategies. In offshore stages, as a result of space imperatives, minimal physical and substance frameworks are utilized. Unfortunately, current advances cannot dislodge these minute suspended oil particles. In addition, natural pretreatment of wastewater can be financially expensive. As high salt fixation and varieties of influent qualities have an impact on PW, it is suitable to fuse a physical treatment (e.g., film) to refine the material. Hence, future research endeavors are concentrating on the streamlining of flow innovations, utilization of consolidated methodology, organic treatment of delivered water, and review of reuse and release limits.

Agricultural wastewater, which comes from all animal farms and food processing, requires unique treatment before disposal or reuse [28]. Untreated agricultural wastewater results in pollution of groundwater, rivers, and lakes, thereby disrupting ecosystems and resulting in a

chain of negative effects. However, with proper treatment and filtration, this wastewater can become a valuable resource. Primary treatment involves separating solids from the liquids and producing “sludge.” The secondary treatment removes contaminants and dissolved solids from the effluent. Ultraviolet light, specialized enzymes, and microbes are often used for further treatment [29, 30]. After which, the “safe” water is returned to a waterway (ocean or river) or reused in agriculture [31]. Thus, treated wastewater can be reused in a sustainable fashion.

Where efficient irrigation methods and collection of run-off are in place, there is little wastewater [tailwater] to be treated for reuse. However, when bountiful tailwater is available, it often contains large amounts of salt and nutrients which makes it non-permissible for irrigation [31]. Innovative effluent treatment permits water reuse for irrigation and animal needs, making the “sludge” and subsequent effluent suddenly valuable. Additionally, collecting and reusing tailwater can benefit a farm through fertilization, and it can protect the environment by avoiding salt and nutrient discharge. Thus, utilizing tailwater and food processing wastewater could be profitable for farmers and positive for our environment.

## 1.2. Membrane fouling

Most membrane technologies experience reduction in performance as a result of various types of fouling. Therefore, designing and investigating membranes to combat fouling is imperative in creating proficient systems. Membrane fouling is the accumulation of unwanted matter such as colloids, salts, and microorganisms during the water purification process. Foulants accumulating on the surface reduces the water flow either temporarily or possibly permanently. Unfortunately, this is a common problem, and these foulants deteriorate and increase the ineffectiveness of the system.

During mass transport, various aspects lead to adsorption of particles within and onto the membrane surface, causing membrane fouling [22]. Contaminated feed water results in compounds and unwanted material adhering to the membrane, resulting in fouling, which is a major problem for most membrane-based systems and often results in a decline in flux [23]. Therefore, minimizing fouling is the key to optimal membrane operation and keeping costs down. Depending upon the polymer utilized for membrane fabrication, additional characteristics can be optimized to prevent fouling. Regardless of the membrane system, biofouling is a long-term problem [32]. All types of fouling (biofouling, organic, colloidal, and scaling) can be damaging [32]. It has been noted that FO is less likely to foul and less complicated than pressure-driven membrane processes like RO [23, 32]. This is because applied hydraulic pressure causes compact foulant layers, which diminish the effectiveness of cleaning the membranes.

Biofouling is considered to be the most difficult and detrimental to water filtration processes and decreases the durability of membranes. Therefore, membranes that are resistant to the accumulation of microorganisms are a necessity for water purification. Ultimately, biofouling causes higher than necessary energy consumption, deterioration of system performance, and water production. Due to the aforementioned issues, it is technologically essential to find efficient methods to minimize membrane biofouling. Studies have shown that FO membranes

are more effective in preventing foulant permeation into the draw solute and reducing fouling in the downstream RO membrane [23].

Organic foulants are dominant and precursors to biofouling when using membrane bioreactor (MBR) for wastewater treatment [22, 33]. Therefore, biofouling can be prevented by controlling the organic matter. Hydrophobic and hydrophilic polysaccharides and transphilic organic macromolecules are all found in the feed water and may lead to organic fouling. Of these examples, polysaccharides are three times more likely than other humic acid contaminants to cause fouling [33].

### 1.3. Membrane selection

Material selection for membrane fabrication is significant in developing a system with optimal flux, as flux decline is directly connected to membrane fouling. Regardless of the polymeric material, asymmetric membranes are preferred during liquid separation due to their thin top layer on top of a porous support layer. FO asymmetric membranes consist of a dense active layer and a loosely bound support layer. The dense top layer is selective and the large pores in the support layer reduce hydraulic resistance [34]. Thin-film composite (TFC) and polysulfone are currently the most widely used materials for membrane fabrication due to their stability and high-pressure tolerance. However, Poly [vinyl alcohol] (PVA) hydrogels have been shown to be a suitable membrane used for water treatment, and PVA is an excellent surface modifier. Their hydrophilicity, water permeability, and anti-fouling potential make them ideal candidates in the further development of composite membranes [35, 36]. Research continues to investigate ways to optimize PVA hydrogel membranes based on their degree of polymerization and incorporation of nanoparticles [37]. Furthermore, studies have proven that ideal membranes should have high water permeability, selectivity, and stability [14].

### 1.4. Concentration polarization

As many are investigating FO for wastewater treatment and desalination, one of the major weaknesses of FO is internal concentration polarization (ICP). The configuration of the membrane contributes to the aforementioned fouling possibility and other complications such as ICP which minimized flux efficiency [33]. Traditionally, the support layer faces the feed in normal mode and faces the active layer in the reverse mode. The inability of the salt to pass easily through the active layer results in a concentration increase within the support layer. Amid the process, fouling such as scaling contributes to concentrative ICP [22, 33]. In the normal mode, the support layer diminishes water transport hydraulic resistance, and the solute freely enters, leading to minimum ICP [38]. Just as fouling leads to lower water flux, ICP within asymmetric thin-film composite (TFC) FO membranes does the same. Contrarily, in reverse mode, the active layer faces the feed solution contributing to ICP. The concentration is increased in the support as the active layer prevents the passage of salt. Thus, ICP greatly reduces the driving force for transport. However, a thin low porosity support minimizes ICP [33] and surface modifications, such as coating with another polymer, has been one of the most effective methods [21]. Studies have been conducted to improve membrane design for new-generation FO membranes and mitigate the ICP effect. Researchers have explored membrane

structures to prevent salt leakage and minimize ICP in FO [39]. Altering phase inversion fabrication protocol by examining different casting substrate, consequently, results in an open structure with increased porosity in the middle support layer. During desalination, the FO system showed decreased salt leakage with mitigated ICP [21]. The ICP and ECP (external concentration polarization) structural value of the double dense-layer membrane is much smaller than those reported in the literature [21]. Moreover, lower CP values were seen after an intermediate solvent/water immersion was performed before complete immersion in water [39]. Additionally, Tang et al. [33] investigated ICP and fouling during humic acid filtration. They reported that despite initial ICP, the active facing orientation resulted in stable flux in contrast to flux diminution when facing foulant humic acid feed water.

## 2. Pressure retarded osmosis

Most water purification processes are known to consume energy. However, using the salinity differences between two bodies of water, pressure retarded osmosis (PRO) generates power. PRO is based on membrane technology similar to FO but results in sustainable osmotic power energy. During PRO, additional back pressure is applied to the draw solute, creating chemical potential between seawater and fresh water. As a result, electricity is produced from the conversion of flux into mechanical energy [22], and the net flux is similar to FO in the direction of the DS [40]. Unfortunately, membrane fouling consequently reduces the permeate flux and osmotic power generation, thus increasing overall cost similar to other membrane technologies. Research has been conducted on different quality feed waters to identify the main foulants on the surface in the PRO processes, and silica has been shown to cause severe scaling [41]. Again, structural parameters, material choice, pH of FS and/or DS played a critical role in mitigating IC of silica scaling [41]. Furthermore, organic and inorganic salt water was used to investigate cleaning methods to resolve fouling issues [32]. Using salt water as the DS, iron, aluminum, calcium, sodium, and silica were the inorganic foulants discovered [32]. Also, humic substances, polysaccharides, and proteins were the organic foulants identified [32]. Sequential acidic and basic cleaners were proven to be successful with a flux recovery above 95% [32]. PRO processes and consequently osmotic power generation can be enhanced by decreasing membrane fouling via chemical cleaning [32].

## 3. Summary

In summary, many researchers have compared FO, PRO, and RO as shown in **Figure 1** [22]. The most noted comparisons are the necessary pressure difference, fouling tendencies, and application. All three systems have advantages but require necessary improvements for expansion of utilization in various applications. Although fouling is a challenge for membrane technologies, research has demonstrated various ways to diminish its effects on flux [22, 32, 41]. With the increasing water demands, FO is certainly a viable option to meet the water and energy challenges of a growing global population as PRO has the potential to be widely used for sustainable energy. With polymer chemistry and membrane innovations, FO will advance for continuous use in producing safe water for irrigation, pharmaceuticals, and human

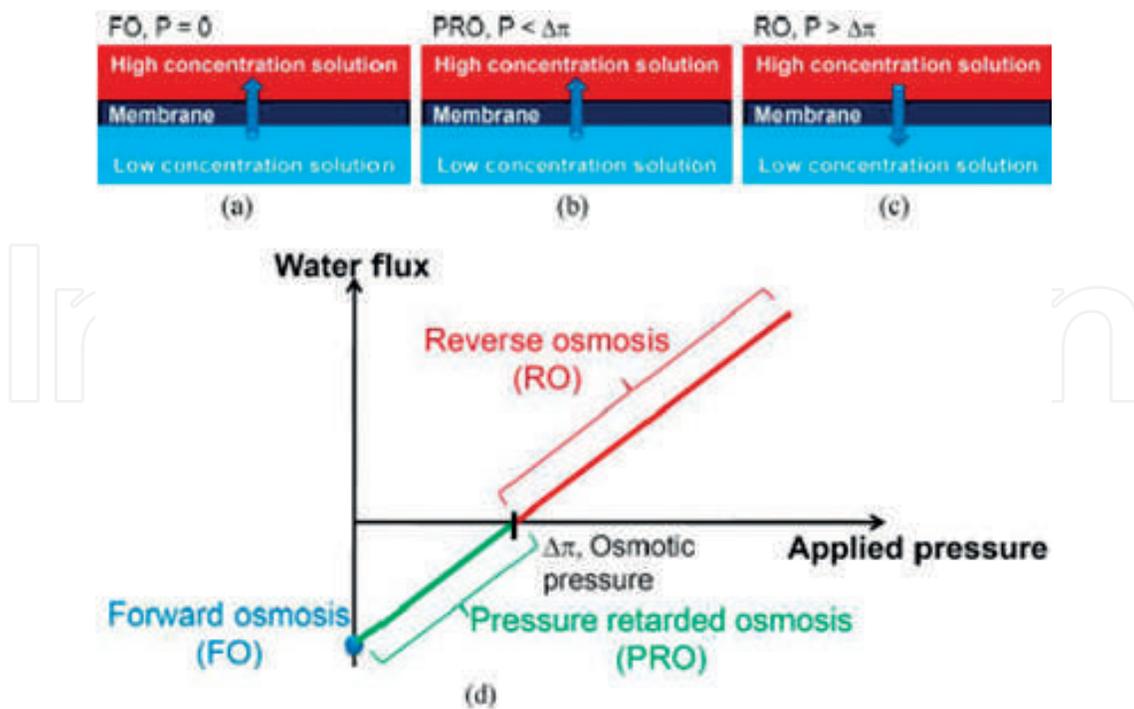


Figure 1. Illustration of FO, PRO, and RO processes [22].

consumption. This book will further discuss the headway in osmotically driven membrane processes (ODMP) research, findings, and contributions to membrane processes.

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# Forward Osmosis Membranes – A Review: Part I

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## Abstract

Forward osmosis (FO) is a technical term describing the natural phenomenon of osmosis: the transport of water molecules across a semi-permeable membrane. The osmotic pressure difference is the driving force of water transport, as opposed to pressure-driven membrane processes. A concentrated draw solution (DS) with osmotic pressure draws water molecules from the feed solution (FS) through a semi-permeable membrane to the DS. The diluted DS is then reconcentrated to recycle the draw solutes as well as to produce purified water. As a major disadvantage, nature of FO membranes (asymmetrical structure) causes international concentration polarization (ICP) which promotes the decrease in water flux. Therefore, the number of studies related to improving both active and support layers of FO membranes is increasing in the applications. The purpose of the chapter is to bring an overview on the FO membrane manufacturing, characterizing and application area at laboratory or full scales. This chapter is published in two parts. In the first part, which appears here, the overview of membrane technologies and the definition of forward osmosis process are stated. The manufacturing methods of support and active layers forming FO membranes are described with common and/or new modification procedures.

**Keywords:** forward osmosis, water flux, reverse salt flux, active layer, support layer, thin film composite, structural parameter, porosity, internal concentration polarization

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## 1. Introduction

Membrane separation processes have been widely applied for many years in environmental, industrial applications, and domestic use such as water/wastewater treatment, desalination, specific industrial purposes and energy recovery. Among the concentration-driven processes, FO

has recently attained many attractions due to its advantages such as less energy requirement, lower fouling tendency or easier fouling removal and higher water recovery. However, all drawbacks of FO process such as; (i) membrane fouling originated from ICP, (ii) lower flux, and (iii) reverse salt diffusion (RSD) limit the performance of the FO applications in environmental studies. Moreover, “necessity of concentrate management” and “meeting of discharge standards with high amounts of product water” oblige the developing new processes, membrane materials or modifications and finding new DS. In recent studies, developing new FO membranes by optimization of thickness, porosity, tortuosity of active/support layer of FO membrane to increase water flux and decrease ICP are mainly focused.

FO is a technical term describing the natural phenomenon of osmosis: the transport of water molecules across a semi-permeable membrane. The osmotic pressure difference is the driving force of water transport, as opposed to pressure-driven membrane processes. A concentrated DS with osmotic pressure draws water molecules from the FS through a semi-permeable membrane to the DS. The diluted DS is then reconcentrated to recycle the draw solutes as well as to produce purified water. As the driving force is only the osmotic pressure difference between two solutions which means that there is no need to apply an external energy, this results in low fouling propensity of membrane and minimization of irreversible cake forming which are the main problems controverted by membrane applications -especially- in biological treatment systems (e.g. FO-MBR). However, nature of FO membranes (asymmetrical structure) causes ICP which promotes the decrease in water flux. Therefore, the number of studies related to improving both active and support layers of FO membranes is increasing in the applications.

In this first part of chapter, advantages of FO over conventional membrane processes and main drawbacks originated from the nature of FO membranes are thoroughly stated by considering both review and research articles in the available literature. The book chapter consists of three main titles firstly including introduction section, the second of which states a literature survey on early definitions of diaphragm, membrane and selectivity phenomenon by considering about last two centuries. Basic principles of FO phenomenon is also expressed in this part. However, the special aspects of FO process are discussed in the third part in view of FO membrane properties. The water flow is mainly determined by the support layer, while the selectivity is by the active layer of FO membrane. Therefore, both support and active layer designing are overemphasized by addressing new materials, manufacturing methods and modification steps to overcome the main challenges of FO processes such as low water flux and concentration polarization phenomena causing the membrane fouling.

## **2. General aspects of membrane processes**

### **2.1. Membrane technology**

Systematic studies of the membrane phenomenon can be traced to the eighteenth century philosopher scientists. For example, Abb'e Nolet prepared an 'osmosis' word in 1748 to describe

water permeability through a diaphragm. Through the nineteenth century and beginning of the twentieth century, membranes were not used for industrial or commercial purposes, but they were used as laboratory tools to develop physical/chemical theories. For example, the measurement of solution osmotic pressure by membranes by Traube and Pfeffer was used by van't Hoff in 1887 to improve the limit law, which describes the behavior of ideal diluted solutions; this work led directly to the van't Hoff equation. At the same time, Maxwell and others used a perfectly selective semipermeable membrane concept in the development of the kinetic theory of gases [1].

Early membrane researchers have experimented with all sorts of diaphragms available for themselves, such as pigs, cattle or fish and sausage covers made of animal guts. Later, nitrocellulose membranes were preferred because they could be reproducibly produced. In 1907, Bechhold developed a technique for preparing nitrocellulose membranes of a graded pore size determined by the bubble test [2]. Other early workers, the technique of Bechhold and were introduced into the market of microporous nitrocellulose membranes at the beginning of the 1930s [3–5]. In the following 20 years, this early microfiltration (MF) membrane technology has expanded to other polymers, especially cellulose acetate. Membranes found their first important practice in drinking water testing at the end of the Second World War. Drinking water sources serving large communities in Germany and elsewhere in Europe were destroyed and filters were urgently needed to test water safety. The research effort to develop these filters, backed by the US Army, was later exploited by Millipore Corporation, the first and largest producer of US MF membranes. By 1960, elements of modern membrane science were developed, but membranes were used only in a few laboratories and small, specialized industrial applications. An effective membrane industry was not available and the total annual sales of the membranes for all industrial applications probably did not exceed US \$ 20 million in 2003. There were four problems that prevented membranes from being widely used as a separation process: they were too unreliable, too slow, too unselective and too expensive. Solutions for each of these problems have been developed over the past 30 years and membrane separation systems have become more common [1].

The first discovery of the conversion of membrane separation into an industrial process from a laboratory appeared with defect-free, high-flux anisotropic reverse osmosis (RO) membranes produced by the Loeb-Sourirajan process at the beginning of the 1960s [6]. These membranes consisted of an ultra-thin, selective surface on a much thicker but more permeable microporous support providing mechanical strength. The flux of the first Loeb-Sourirajan RO membrane was 10 times higher than that of any available membrane, and this performance made the RO potentially a practical method for desalinating water. Loeb and Sourirajan's work and large-scale research and development by the US Department of Interior Office of Saline Water (OSW) have been a pioneer in the commercialization of RO and this has been a major factor in the development of MF and ultrafiltration (UF) membranes. In addition, the development of electro dialysis was supported by OSW funding. With the development of these industrial applications of membranes, the development of

-especially- artificial kidneys, has been provided for medical separation procedures. Kolff et al. [7] demonstrated the first successful artificial kidney in Holland in 1945. It took about 20 years for technology to be applied to large-scale works, but these developments were completed in the early 1960s. Since then, the use of membranes in artificial organs has become an important life-saving procedure [1].

Currently, more than 800,000 people are protected with artificial kidneys and about 1 million of people who have undergone open heart surgery every year through a possible procedure by developing a membrane blood oxygenator. The sales of these devices easily exceed the total industrial membrane separation market. Another important medical application of membranes is for controlled drug delivery systems. An important figure in this area was Alex Zaffaroni, who founded Alza, a company dedicated to developing these products in 1966. Membrane techniques developed by Alza and his competitors are widely used in the pharmaceutical industry to improve efficacy and safety of drug delivery. Significant stages were recorded in the membrane technology of 1960–1980 period. Using the original Loeb-Sourirajan technique, other membrane production methods including interface polymerization and multilayer composite casting and coating were developed to produce high performance membranes. Membranes produced using these methods and containing thin layers of 0.1  $\mu\text{m}$  or less are now produced by many companies. Along with the membrane type, membrane modules have been developed by working on the packing volume and the number of studies for increasing the membrane stability has increased. In the 1980s, large-scale installations involving MF, UF, RO and electrodialysis began to become widespread all over the world [1].

## 2.2. Conventional membrane processes

RO is primarily used to remove salts from brackish water or seawater while it can reject synthetic organic compounds. One of the latest developed membrane process, nanofiltration (NF), is used to soften fresh water and clear disinfection by-products (DBP) precursors. Electrodialysis is used to demineralize brackish and sea water and to soften fresh water. UF and MF are used to remove turbidity, pathogens and particles in fresh water. In the broadest sense, a membrane, a common element of all of these processes, can be defined as any barrier to the flow of suspended, colloidal or dissolved species in any solvent. Applicable size ranges for membrane processes are shown in **Figure 1**. Generally, the cost of membrane processing increases when the size of the solute is reduced. The ionic range in **Figure 1** includes potable water solubles such as sodium, chloride, total hardness, maximum dissolved solids, and smaller DBP precursors. Macromolecular range includes large and small colloids, bacteria, viruses, and colors. The fine particle range includes particles that produce larger turbidity, most suspended solids, cysts, and larger bacteria. Membrane processes normally used in the ionic range can remove macromolecules and fine particles, but are not as cost effective as larger pore membranes due to some operational problems [9]. The comparison of the membrane properties with each other is given in **Table 1**.

RO is the tightest membrane process in liquid/liquid separation. In principle, water is the only substance passing through the membrane; essentially all dissolved and suspended material is rejected. The RO membranes with much larger pores are sometimes confused with

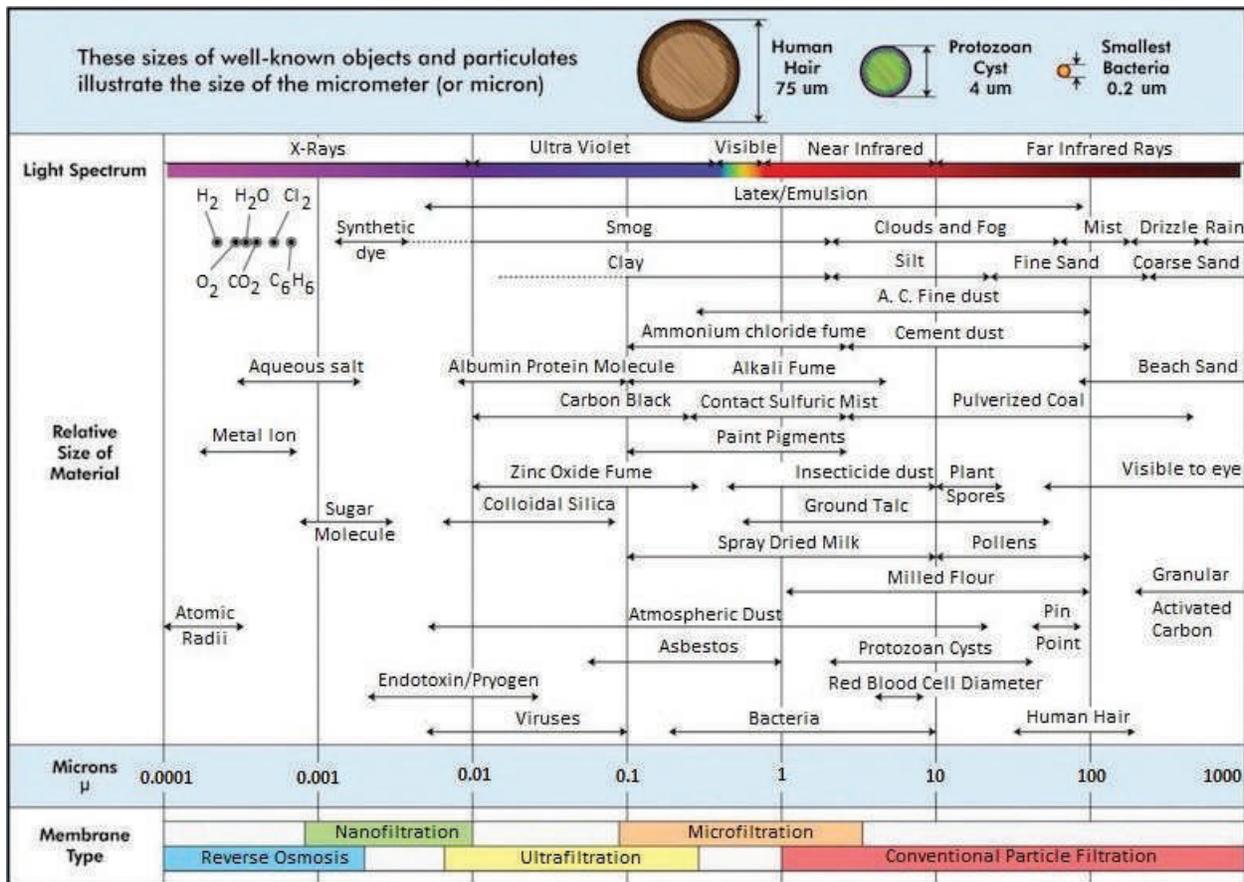


Figure 1. Membrane pore size compared with molecules, bacteria and virus [8].

NF membranes. True NF rejects ions with more than one negative charge, such as only sulfate or phosphate, while passing single charged ions. NF also rejects uncharged, dissolved materials and positively charged ions according to the molecular size and shape. Finally, the sodium chloride (NaCl) rejection varies from 0 to 50%, depending on NF and the rejected feed concentration. In contrast, “loose RO” is an RO membrane with reduced salt rejection. Such membranes are highly effective for a range of applications where moderate desalination is acceptable and, therefore, the operating pressure and power consumption are significantly reduced. Therefore, the costs are reduced in cases where complete desalination is not required.

UF is a process in which all low molecular weights compounds (LMWC) are freely permeable, while the high molecular weights compounds (HMWC), such as proteins, and suspended solids are rejected. Therefore, none of the mono- and di-saccharides, salts, amino acids, organics, inorganic acids or sodium hydroxide are rejected. Microfiltration (MF) is ideally a process where only suspended solids are rejected, and even proteins pass free through the membrane.

A wide range of products using membranes, but water desalination uses more than 80% of all membranes having ever been sold. The remaining 20% is used for -mostly- milk processing, while the rest is sold for use with many different liquids. Some liquids are waste products and some of which are very expensive pharmaceutical products. **Table 2** lists some typical applications and as seen in the table the permeate as well as the concentrate can be the desired product [10].

	<b>Reverse osmosis</b>	<b>Nanofiltration</b>	<b>Ultrafiltration</b>	<b>Microfiltration</b>
Membrane	Asymmetrical	Asymmetrical	Asymmetrical	Symmetrical Asymmetrical
Thickness	150 $\mu\text{m}$	150 $\mu\text{m}$	150–250 $\mu\text{m}$	10–150 $\mu\text{m}$
Thin film	1 $\mu\text{m}$	1 $\mu\text{m}$	1 $\mu\text{m}$	
Pore size	<0.002 $\mu\text{m}$	<0.002 $\mu\text{m}$	0.2–0.02 $\mu\text{m}$	4–0.02 $\mu\text{m}$
Rejection of	HMWC, LMWC Sodium chloride Glucose Amino acids	HMWC Mono-, di- and oligosaccharides Polyvalent neg. ions	Macromolecules, Proteins, Polysaccharides Vira	Particles Clay Bacteria
Membrane material(s)	CA Thin film	CA Thin film	Ceramic PSO, PVDF, CA Thin film	Ceramic PP, PSO, PVDF
Membrane module	Tubular, Spiral wound, Plate-and-frame	Tubular, Spiral wound, Plate-and-frame	Tubular, Hollow fiber Spiral wound, Plate-and-frame	Tubular, Hollow fiber
Operating pressure	15–150 bar	5–35 bar	1–10 bar	<2 bar

**Table 1.** Comparing membrane processes [10].

### 2.3. Forward osmosis process

van't Hoff's semipermeable membrane, which he assumed to promote the dilution of aqueous solution theory, is a permeable barrier to water (solvent), which is completely impermeable to dissolved solutes. For this reason, removal of the solvents results in a model barrier for all membrane filtration processes where the solutions are retained (concentrated). Like all joining properties, osmotic effects are limited to liquid solutions. Since we know nature is a watery system, the following solvent is water. When pure water and a random aqueous solution come in contact with the environment through a semi-permeable membrane, pure water is "drawn" into the solution, as if to dilute it: Osmosis. As is well known, osmosis is extremely important for the functioning of life when understood as a transport phenomenon at the molecular level. Live cell walls are osmotic barriers with improved selectivity towards inorganic and organic solutes ("biological membranes"). The direction of osmotic water transport, irrespective of the nature of the solution, indicates that the solution has a lower free energy (potential) than pure water. Specifically, the effectiveness of the solvent must be reduced by the effect of the solute, since the model barrier is assumed to communicate only through solvent [11].

Forward osmosis, an evolving separation/desalination process, has received increased interest in both academic research and industrial development in the past decade [12]. In FO, a semi-permeable membrane is placed between two solutions of different concentrations: a concentrated DS and a more dilute FS. Using the osmotic pressure differential to provide water permeation through the membrane, FO may respond to some of the deficiencies of hydraulic pressure driven membrane processes such as RO [13].

		Permeate	Concentrate
RO	Dyeing effluent	Clean water	BOD, salt, chemicals, waste products
	Water	Low salinity water	Salty water
	Whey	Low BOD permeate	Whey concentrate
	Antibiotics	Salty waste product	Desalted, concentrated antibiotics
NF	Dyeing effluent	Clean, salty water	BOD/COD, color
	water	softened water	waste product
	Whey	Salty waste water	Desalted whey concentrate
	Antibiotics	Clarified fermentation broth	Waste product
	Bio-gas waste	Clarified liquid for discharge	Microbes to be recycled
	Carrageenan	Waste product	Concentrated carrageenan
	Enzymes	Waste product	High value product
	Milk	Lactose solution	Protein concentrate for cheese production
UF	Oil emulsion	Oil free water (<10 ppm)	Highly concentrated oil emulsion
	Washing effluent	Clarified water	Dirty water (waste product)
	Water	Clarified water	Waste product
	Whey	Lactose solution	Whey protein concentrate
	Xantan	Waste product	Concentrated xantan

Table 2. Type of membrane process for several products (the shaded area representing the main product) [10].

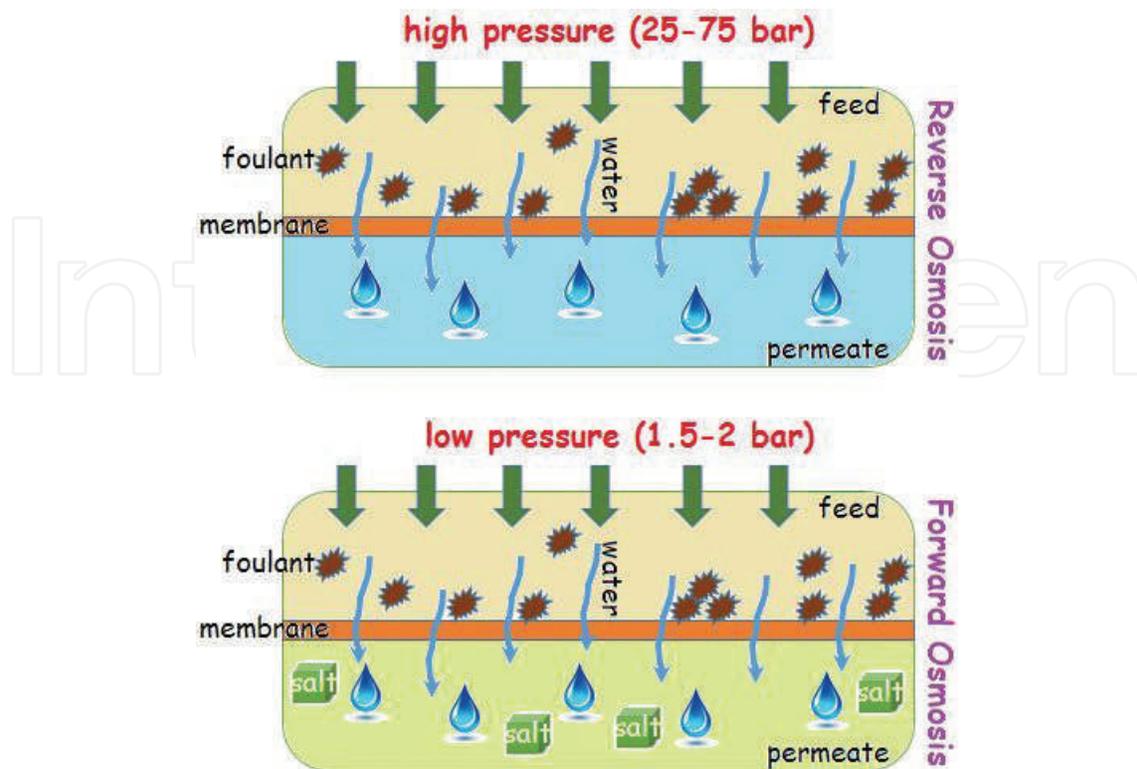
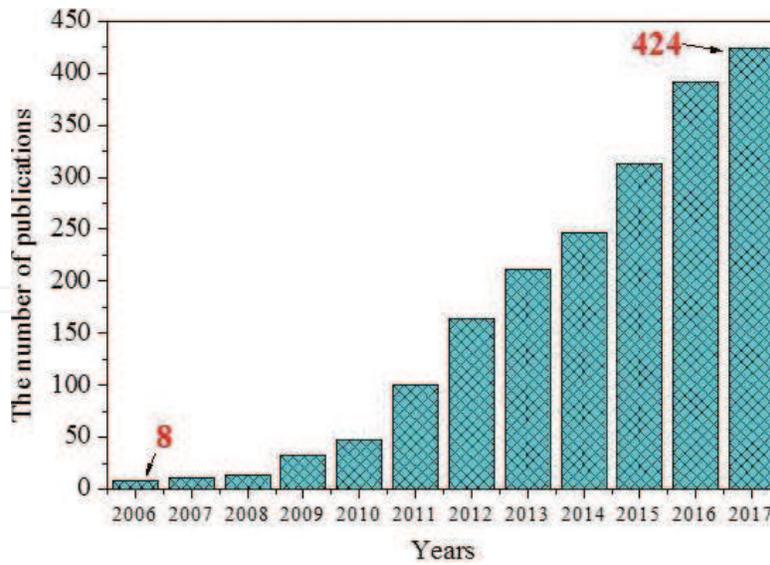
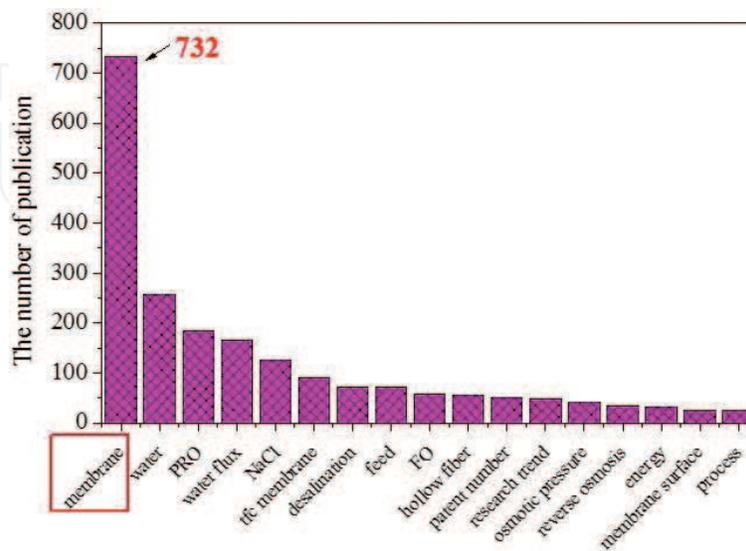


Figure 2. Illustration of comparison between FO and RO processes [16].



**Figure 3.** Annual number of publications on FO since 2006 until fourth quarter of 2017 (retrieved from science direct database search) (updated and adapted from Eyvaz et al. [16]).

The transport of water molecules from a semipermeable membrane to the concentrate/saline solution on the other side of the membrane is referred to a technical term as forward osmosis. Contrary to conventional pressure-driven membrane processes, no pressure is applied to the side of water or concentrated solution. The difference in osmotic pressure between the aqueous medium on both sides of the membrane serves as the driving force to transport the water. [14]. Concentrated solution (DS), which pulls water molecules, is diluted during the process. The diluted DS is then re-concentrated in order to separate the water from the DS with a suitable further process. Where appropriate, the re-concentrated solution can be used again as DS. [15]. The FO process is shown in **Figure 2**. The main advantages of FO are it is not operated



**Figure 4.** The number of publication about FO studies different research topics since 2006 until fourth quarter of 2017 (retrieved from science direct database search) (updated and adapted from Eyvaz et al. [16]).

under any hydraulic pressure, that a wide range of pollutants can be rejected at a high level and have lower irreversible pollution than pressure-based membrane processes [17].

As a method for water desalination, FO has been investigated for about four decades [18] and many researchers have found that (i) the selection or development of (new) membrane materials [19, 20], (ii) determining the suitable DS [21] The understanding of the mechanism of pollution [22], (iv) the characterization of concentration polarization (CP) [23]. In these pure academic publications, the FO survey and the increasing tendency of various special topics have been shown in **Figures 3** and **4** for the last 10 years. As seen in **Figure 3**, the number of researchers has been steadily increasing, and recent research has focused heavily on membrane properties and development [16].

### 3. Manufacturing of forward osmosis membranes

Currently used membranes are mostly asymmetric porous membranes [24, 25]. In asymmetric porous membranes, the structure and transport properties change across the membrane thickness. An asymmetric membrane normally consists of a dense layer of 0.1–1  $\mu\text{m}$  thick and supported by a highly porous, 100–200  $\mu\text{m}$  thick support layer [24]. The dense layer provides most of the selectivity for the membrane. The separation properties are determined by the chemical structure, the size of the pores (0.4–1 nm) and the thickness of the skin layer. It is believed that the porous substrate provides mechanical support for the thin and fragile selective layer and has little effect on the separation performance of the membrane. However, recently the effects of the chemical properties of the support layer (eg, hydrophilicity/hydrophobicity) and pore structure (e.g., pore size and porosity) on composite membrane transport have been reevaluated [26–28].

In thin film composite (TFC) membranes, the porous support layer is generally an integrally skinned membrane formed by a non-solvent induced phase separation (NIPS) process. The skin layer is typically formed by either interfacial polymerization (IP) or dip coating followed by crosslinking [24]. The most common thin film chemistry for RO membranes is based on a completely aromatic polyamide (PA) formed by the IP of meta-phenylenediamine (MDP) and trimesoyl chloride (TMC). In contrast, the popular PA NF membranes are formed by IP of piperazine and TMC [29]. It is assumed that the dense selective layer formed by the IP is heterogeneous (20–200 nm) throughout the thickness and is highly cross-linked. The surface properties of a PA film are different from those of the PA dense layer because the polymer density is not uniformly distributed [30]. The PA dense layer is extremely negatively charged because acyl chloride groups are not completely converted to amide during the formation process; however, recent direct titration experiments have demonstrated the presence of both positive and negative fixed charges in the dense layer of composite PA NF membranes [31]. According to [28], Freger and Srebnik suggested that the fixed charge is not uniform and that the film is actually a “sandwich” with two oppositely charged layers [32].

The dense coating layer has been treated as a non-porous film in the past. More advanced identification techniques such as atomic force microscopy, scanning electron microscopy, transmission electron microscopy (TEM), X-ray photoelectron spectroscopy, electron spin resonance, nuclear magnetic resonance (NMR), small angle X-ray scattering, and molecular

dynamics simulations have been used to state the structure of the dense layer. In the literature [28], a highly cross-linked PA skin layer structure with sub-nanoscale pores (0.2–1 nm) and low porosity has been reported [33, 34].

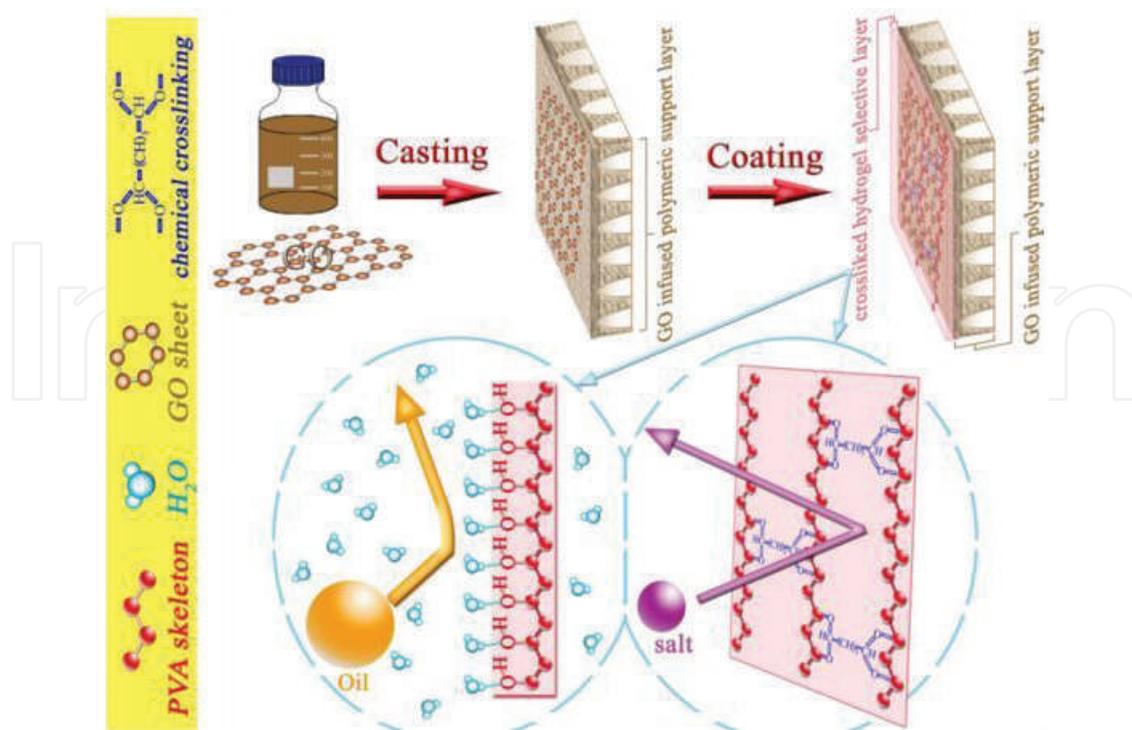
Wang et al. [28] stated that mixed matrix membranes contain both organic and inorganic phases. The first mixed matrix membranes were produced to enhance the performance of the gas separation membranes by providing interconnected flow paths of materials with a high diffusion rate [35]. In aqueous separations, mixed matrix membranes are typically formed of a polymer matrix in which inorganic particles are dispersed. Classically, micron-scale inorganic fillers (eg, zeolites and silicalite) have been added to polymer membranes to create preferential flow paths for rapid transport of certain molecules [36]. When nanomaterials (eg, metal and zeolites nanoparticles) are used as the inorganic filler, these membranes are called nanocomposite membranes [37].

Inorganic particles may be present throughout the thickness of a symmetric or integrally-skinned membrane or exclusively in the coating film of a composite membrane. Theoretically, mixed matrices add an additional degree of freedom to membrane production because the advantages of a particular filler material can be imbued into a bulk membrane material [35]. Mixed matrices have been used to enhance the mechanical and chemical stability of organic membranes and to add specific functionality to the interface of polymer membranes, such as desired degradation, reduced fouling or increased selectivity [38, 39]. Organic and inorganic hybrid membranes are very interested in using it as a new generation of membrane materials for water treatment. According to Wang et al. [28], scientists have begun to use nanoparticles TiO<sub>2</sub> [40], carbon nanotubes [41], zeolites [42], clay [43], nonporous amorphous silica [44] and such as, to increase the water flux.

In another study [45] a new nanocomposite FO membrane is designed to perform oil/water separation and desalination at the same time. This nanocomposite FO membrane consists of an oil-repelling and salt-rejecting hydrogel selective layer on the top surface of graphene oxide (GO) nanosheets grafted into a polymeric support layer (**Figure 5**). This selective layer demonstrates strong underwater oleophobicity, which leads to superior anti-fouling properties under various oil/water emulsions and ICP can be decreased by and can be significantly reduced by GO in view of membrane structural parameter (S) decrease by about 20%. Compared to the commercial FO membrane, this new FO membrane has a markedly low membrane fouling tendency, having higher removal rates for oils and salts (>99.9% in oil and >99.7% in multivalent ions) for treatment of simulated shale gas wastewater (**Figure 6**). These combined advantages will endorse this new FO membrane in the treatment of highly saline and oily wastewater.

Xu et al. [46] reported that the availability of suitable FO membranes is crucial for the development of FO technology. Problems such as high RSD, high concentration polarization (CP) and poor mechanical strength are frequently encountered in FO processes. Meanwhile, although FO tends to exhibit a lower membrane fouling than pressure driven membrane processes, fouling is still the most serious problem that adversely affects FO performance. To overcome these problems, many new FO membranes have been fabricated or ready-made membranes have been modified by means of surface chemistry in recent years [47].

Chung et al. [48] stated that a few comprehensive reviews on FO membrane development are available in the literature [12, 18, 49]. Basically, most FO membranes are fabricated with conventional phase inversion [49] and TFC by IP processes [50]. Each layer of FO membranes have



**Figure 5.** Illustration of the synthetic process and work mechanisms of hydrogel/GO FO membrane [45].

been investigated, but reverse solute diffusions (RSD) tend to be high [51]. Employing hydrophilic materials as substrates in FO membranes is crucial to increase water flux [52]. Recently, TFC-FO-membranes synthesized on nanofiber [53, 54] and multi-bore [20] surfaces with good mechanical properties have also been reported. Future R & D should focus on innovative membranes with low fouling and ICP. Until recently, double-skinned FO membranes with dense RO skin and a loose RO skin, have been promised reduced membrane fouling and ICP [55].

Xu et al. [46] declared that the FO membrane serves as a selective barrier to control the water transport and solute retention to maintain the separation efficiency. The initial attempt to use the RO membrane in the FO process faced with some operational limitations; such as low flux, due to the thick sponge-like substrate and compact support of the RO membrane hindering mass transfer and causing severe ICP within the support layer [28–30]. Hydration Technologies, Inc. (HTI) developed the first commercial FO membranes [56], one of which has a characteristic structure embedding a thin polyester mesh support in cellulose triacetate (CTA) (**Figure 7**). These membranes provide significantly better separation performance than commercially available RO membranes. In addition to the commercial CTA FO membranes, HTI has then introduced the TFC FO membrane to the market; The flux of the spiral element was twice that of existing CTA membranes. This is thought to be a new criterion in future studies on FO membranes [58]. However, FO membranes with superior water permeability and salt rejection are still subjects to be developed for commercialization of FO technology. SEM images of the other some commercial FO membranes are presented in **Figure 8**.

According to the research in literature, it is shown that the adjustment of the sub-layers is of great potential in tailoring PA-TFC membranes. In addition, the flexibility of PA-TFC membrane

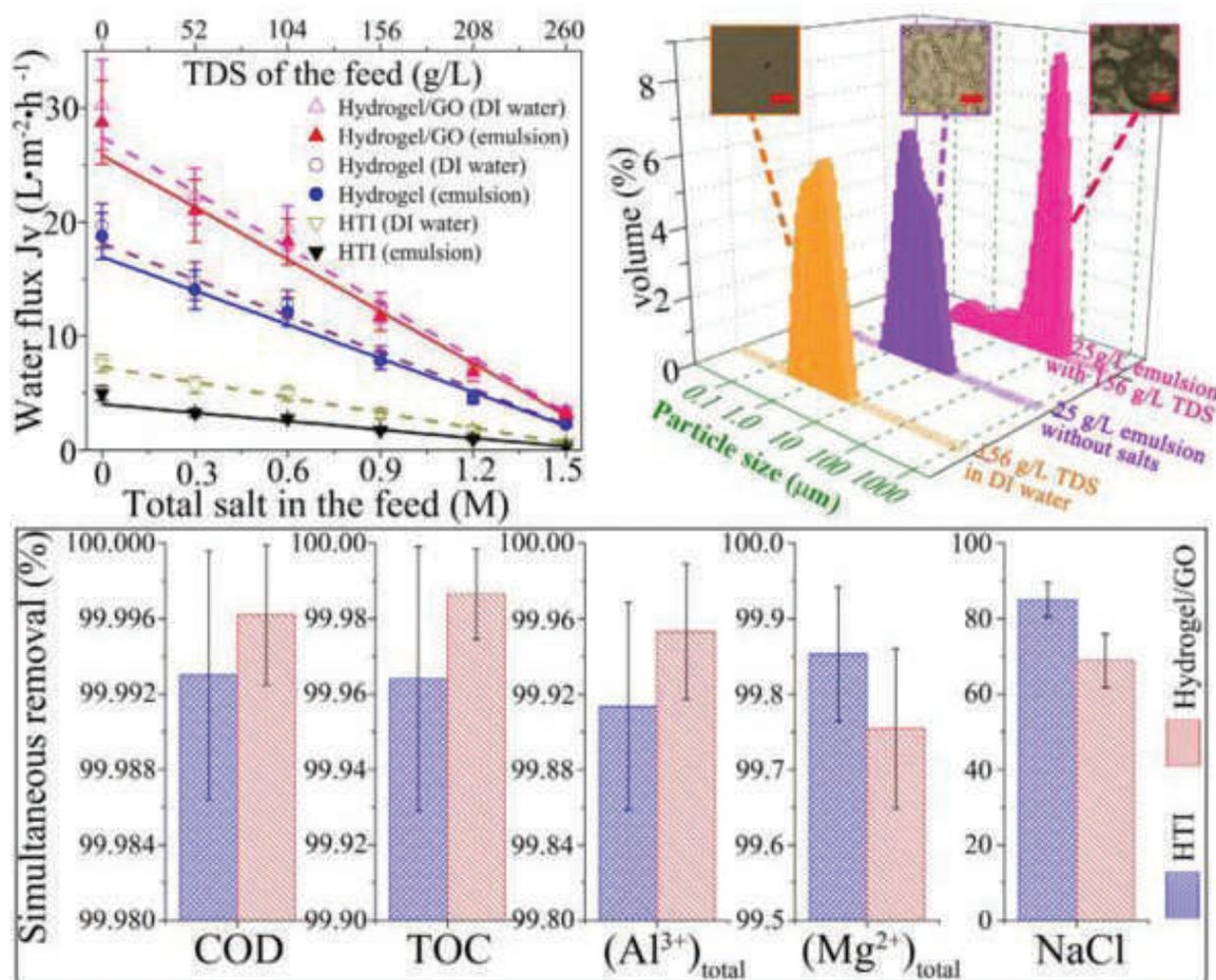


Figure 6. The study of simultaneously deoiling and desalting shale gas wastewaters [45].

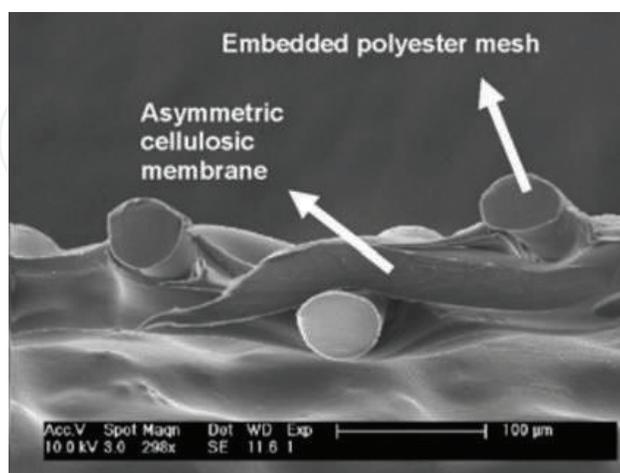
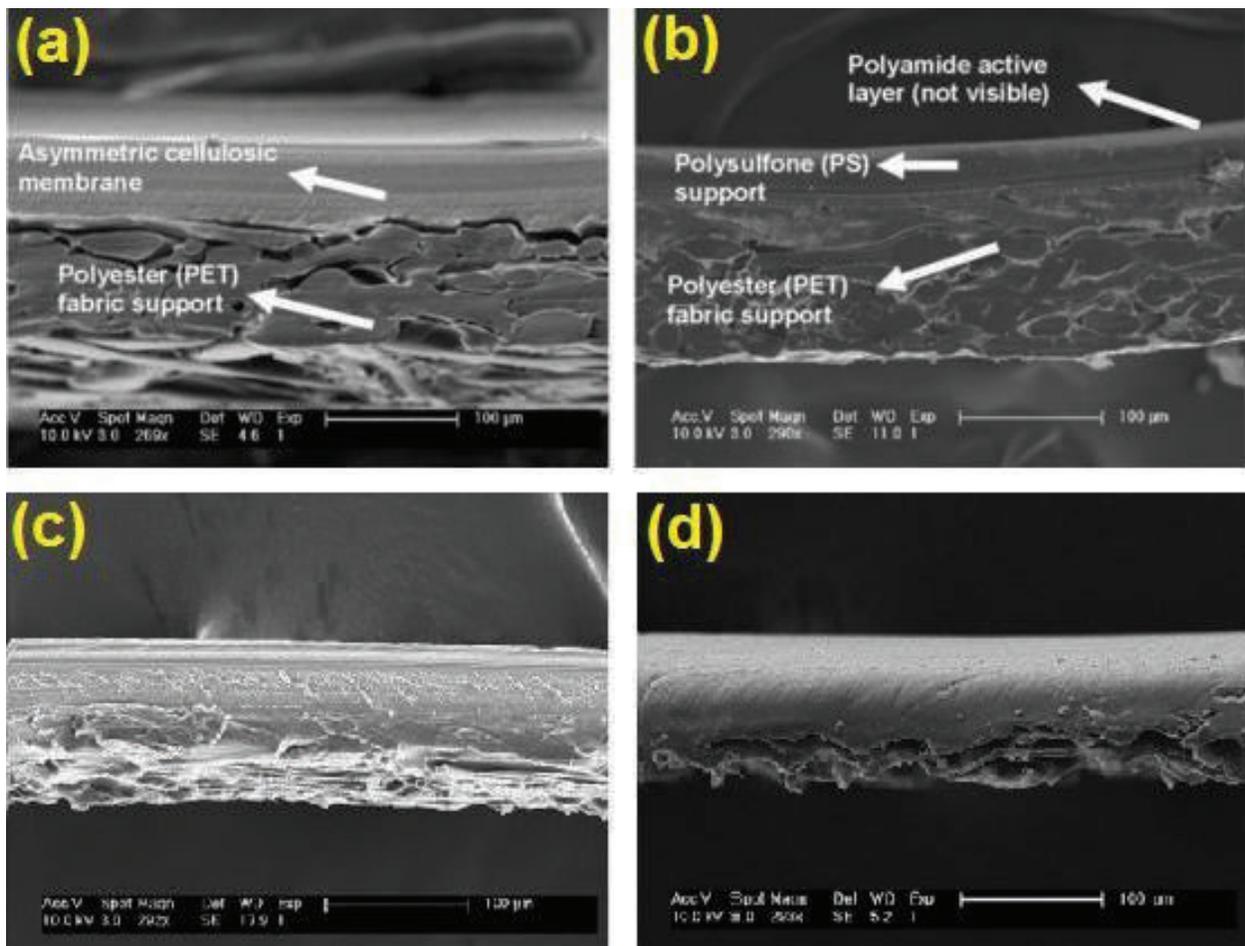


Figure 7. Cross-sectional SEM images of hydration technologies CA (the bar in each SEM image is 100  $\mu m$ ) (adapted from McCutcheon and Elimelech [57]).

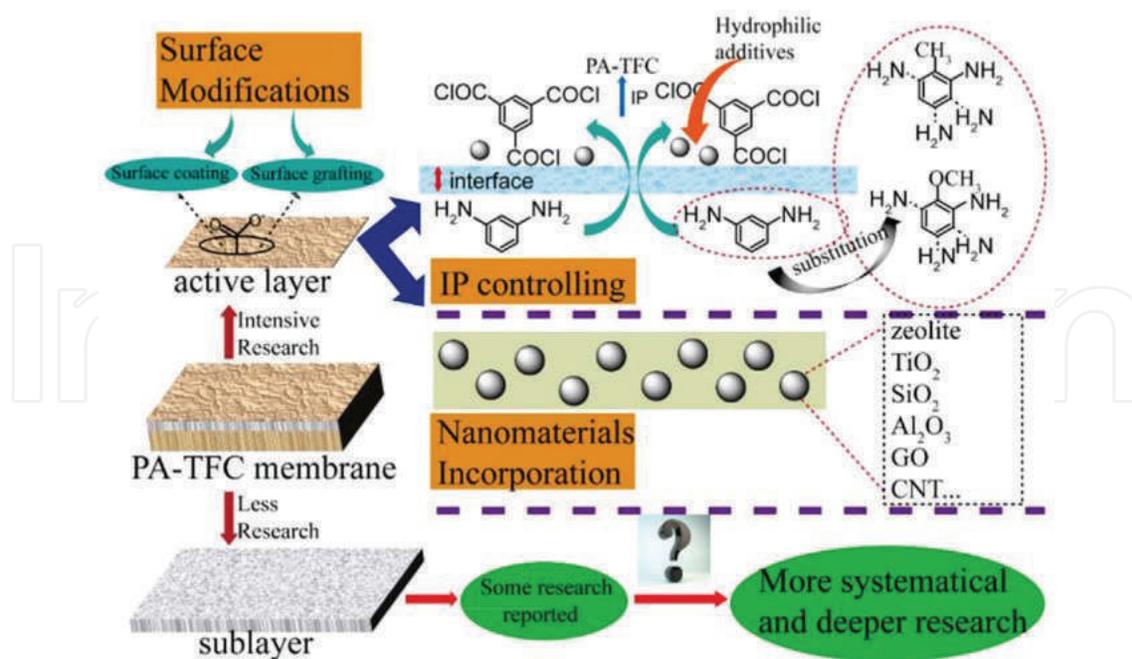


**Figure 8.** Cross-sectional SEM images for the (a) Osmonics CE, (b) Filmtec SW30 XLE and (c) Filmtec SW30 XLE, (d) Osmonics CE membrane with the fabric layer removed (the bar in each SEM image is 100 µm) (adapted from McCutcheon and Elimelech [58]).

structures has a positive effect on the improvement of the sub-layers, since each of the surface active and support layers can be individually constituted for a particular purpose (Figure 9) [59].

### 3.1. Support layer properties and manufacturing techniques

Li et al. [60] have recently reviewed recent researches on polymer and polymer composite membranes for RO and FO processes comprehensively. In one of these studies, a TFC FO flat membrane has a thin selective layer on top of a flat porous polymeric support that is produced by phase inversion with/without a thin nonwoven layer [50]. More recently, nanofiber mats with high porosity have been proposed as a support layer to reduce the ICP to a minimum [53, 54, 61]. Bucky-papers made from CNTs are also being tested as support layer candidates due to their flexibility, strength and high porosity, it is also recommended to investigate other low cost and high porosity materials such as metal oxide nanotubes [62]. Parallel to the studies with TFC flat plate modules, the number of research related to hollow fiber configuration is also increasing due to its advantages such as high packing density and enhanced flow



**Figure 9.** Flexibility in tailoring PA-TFC membranes by adjusting the surface active and support layers individually [59].

pattern and self-supporting structure [20, 63]. Under the same drive force effect, higher fluxes were obtained with hollow fiber membranes than with flat sheet membranes [64].

Similar to the characteristics of the support layers required to produce high performance PA TFC RO membranes, it is desirable that the support layers of the TFC FO membranes have high hydrophilicity, stability and mechanical strength, [65]. In addition, resistance to chemicals, temperature and oxidation, as well as low fouling tendencies, increase the potential use of FO membranes in harsh industrial environmental conditions. [66]. However, the thin support layer, high porosity, and low tortuosity will help reduce ICP [67]. For this reason, some of the research related to FO membranes have focused on support layer fabrication and modification. In these studies, fabrication parameters such as optimization of polymer concentration, solvent composition and functional additives have been considered in the synthesis of the support layer [68]. For example, a simultaneous casting of two polymer solutions with a co-casting technique, a synthesized support layer has played an important role in reducing ICP, improving water flux and reducing reverse salt flux [69]. Furthermore, even the selection of non-woven fabrics underneath the polymeric support layer significantly affects the adhesion of the support layer polymer to this non-woven sheet. For example, selecting a fabric with high tortuosity, large thickness, and low porosity leads to an undesirable decrease in the water flux of the FO membrane. Moreover, the addition of foreign components to the substrate casting solution can help improve the substrate properties. For example, when lignin content is incorporated into the polysulfone (PSf) substrate, the bulk porosity enhanced, shorter diffusion pathway is provided and TFC membrane performance is improved [70]. By using diethylene glycol as a pore forming agent in the PSf/sulfonated poly (ether ketone)/N-methylpyrrolidone (NMP) casting solution, greater porosity and wider pore size distribution were obtained which reduced the resistance of the support layer to the solution and the reverse salt flux was also relatively controlled [65]. Addition of PEG to the preparation of the CAP substrate increased the connectivity

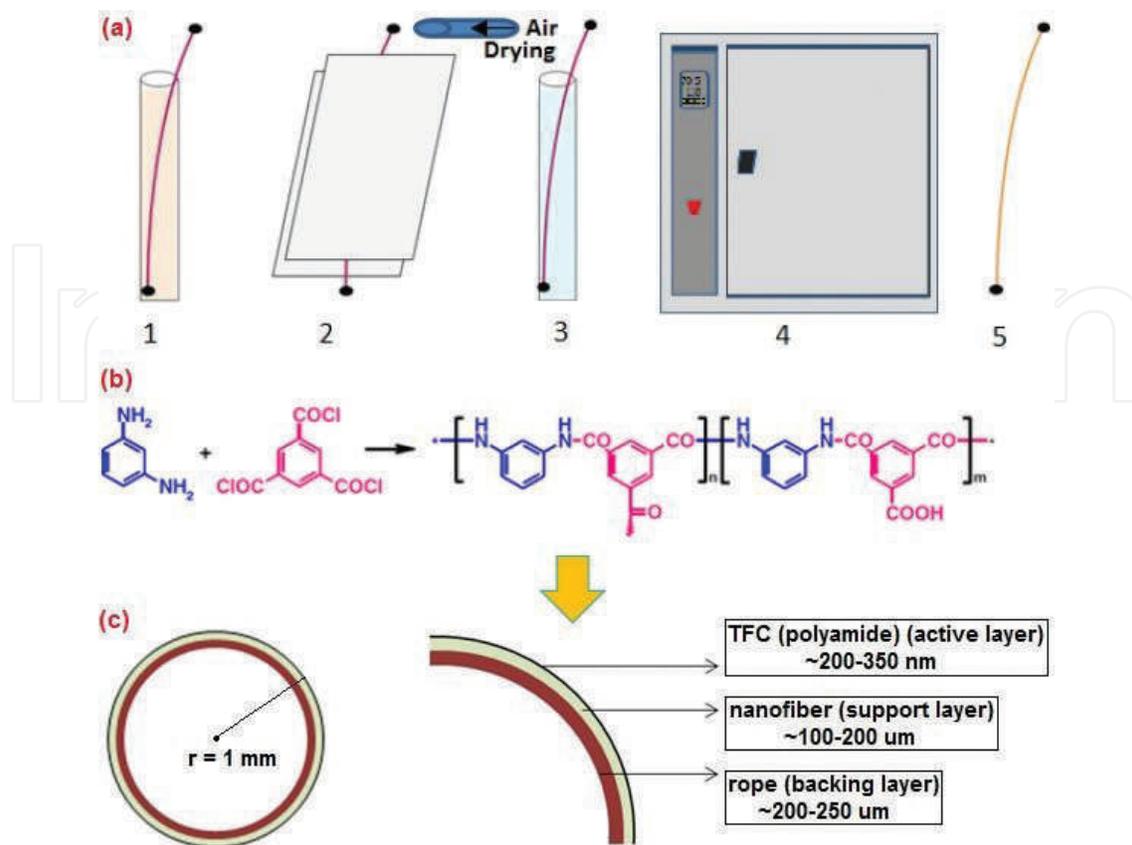
of the pores and prevented macrovoids, as well as positively interacted with the cast glass blade. The resulting support is suitable for high performance TFC FO membrane fabrication since it has a high porosity bottom and a tight top surface [52]. The result is that the TFC membrane with macrovoids (or finger-like structure) support facilitates mass transport and reduces ICP in FO [67]. However, these porous structures may become mechanically weak points in the membrane structure and may, in practice, worsen membrane separation under continuous water flow or backwash conditions [71]. The highly porous support can also increase the difficulty in forming an excellent active selective layer with the necessary mass transport properties [67]. However, the sponge-like structure of small pores surrounded by dense walls may be convenient to form an integral thin active layer and may exhibit better mechanical stability on the finger-like property; however, it increases resistance to mass transfer [64]. Experimental studies suggest that the ideal support with a sponge-like film on a finger-like substrate is very important in the fabrication of high-performance TFC FO membranes [50].

Recently, nanofiber support layers with scaffold-like and interconnected porous structures have been seen as promising alternatives to overcoming the disadvantages of sponge-like structures. The nanofiber supported PA TFC membranes exhibited much lower  $S$  ( $\sim 80 \mu\text{m}$ ) than a commercial HTI FO membrane ( $S = 620 \mu\text{m}$ ) and thus with a low molarity (0.5 M NaCl) DS and a DI water as the FS, it has been observed that the water flux has increased by five times [72].

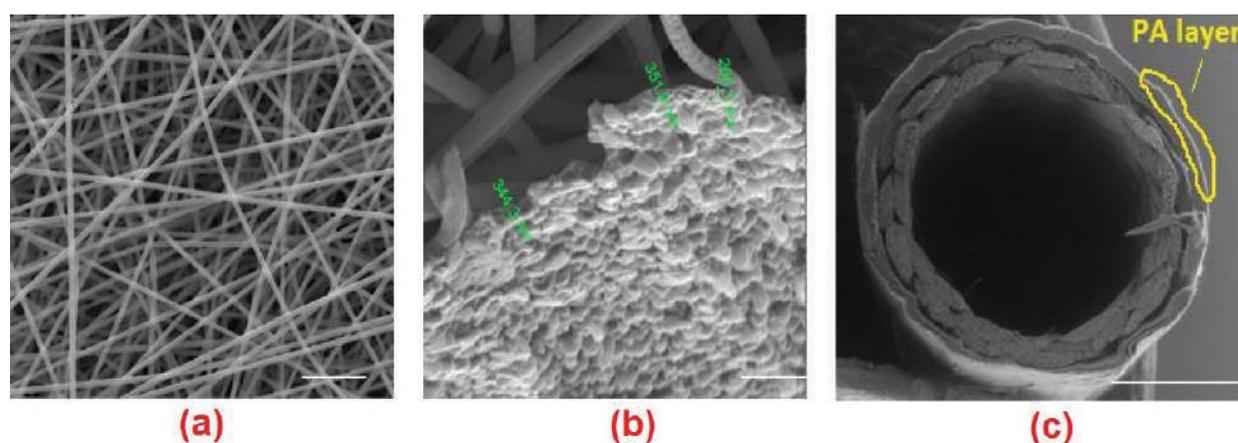
An FO membrane with tubular nanofiber support was manufactured for the first time in the study of Arslan et al. [53]. In the first stage of the manufacturing, the support layer (polyacrylonitrile (PAN) nanofiber) was coated on the hollow braided rope (backing layer) by electrospinning method. In the second step, the active layer called the TFC layer was coated on the formed nanofiber by the IP process. Schematic illustration of FO membrane manufacturing is shown in **Figure 10** and SEM images are presented in **Figure 11**. The TFC layer is the main selective barrier that prevents the transfer of the salt to the diluted side and allows the water molecules to diffusion into the DS side.

According to aforementioned review [60], Han et al. [65] pointed out that the hydrophilicity and support layer thickness are critical parameters in controlling of water transport. It is reported that the TFC membrane with support layer which is completely sponge-like and has a hydrophilic upper surface, provides a higher water flux than a TFC membrane with support layer which is completely hydrophobic and has finger-like structure. In order to prepare or modify the support layer, hydrophilic materials such as sulfonated polysulfone (sPSf), sulfonated copolymer made of polyethersulfone (PES) and polyphenylsulfone (PESU-co-sPPSU), sulfonated poly(ether ketone), poly-dopamine (PDA) or poly(vinyl alcohol) (PVA) have been explored [73–75]. Emadzadeh et al. improved the mass transfer and reduced ICP by applying TFC on the PSF support layer containing  $\text{TiO}_2$ , thus increasing the water flow of the FO membrane [76].

Liang et al. [77] for the first time in the production of TFC-FO membranes, it has been proposed to use vertical porous substrates as a support layer. The addition of acetone in aqueous phase promotes IP on vertical porous substrates. Positron annihilation lifetime analyses indicated that new FO membranes in the study have thicker and dense selective layers than conventional FO membranes with asymmetric substrates. These new FO membranes have a low structural parameter, indicating a greatly reduced ICP effect.

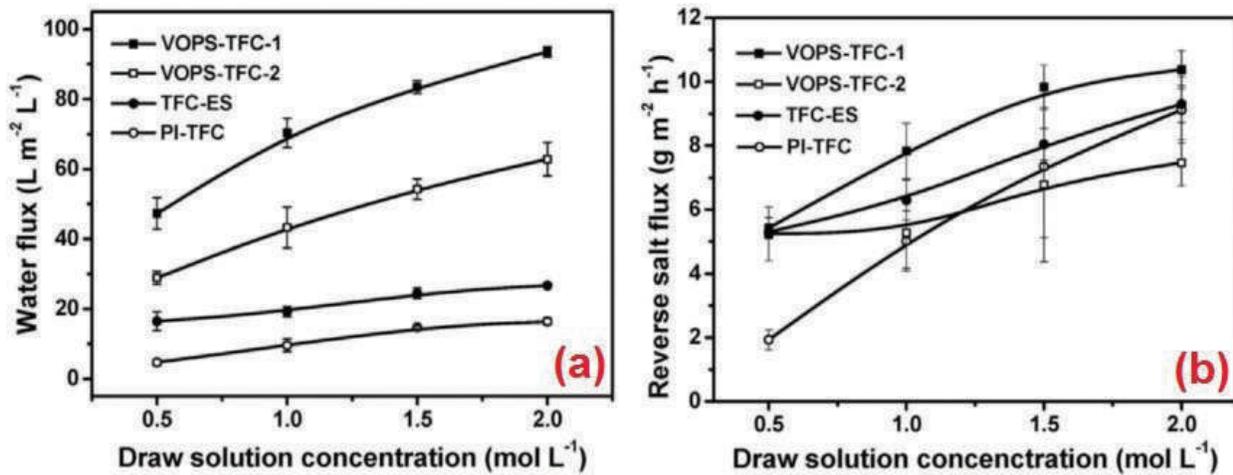


**Figure 10.** Demonstration of tubular nanofiber FO membrane manufacturing steps: (a) Steps of TFC process, 1— immersion of the tubular nanofiber membrane into the MPD solution, 2—air drying, 3—immersion of the tubular nanofiber membrane into the TMC solution, 4—heat treatment, 5— tubular nanofiber FO membrane. (b) Reaction mechanisms of the polyamide formation from MPD and TMC. (c) Schematic representation of cross-section of tubular nanofiber FO membrane [54].



**Figure 11.** SEM images of (a) nanofibers in support layer at magnification of 10,000 $\times$ , (b) gel-like formation of the polyamide layer on nanofibers at magnification of 50,000 $\times$ , (c) cross-section of tubular nanofiber supported FO membrane at magnification of 10,000 $\times$  (adapted from Arslan et al. [53]).

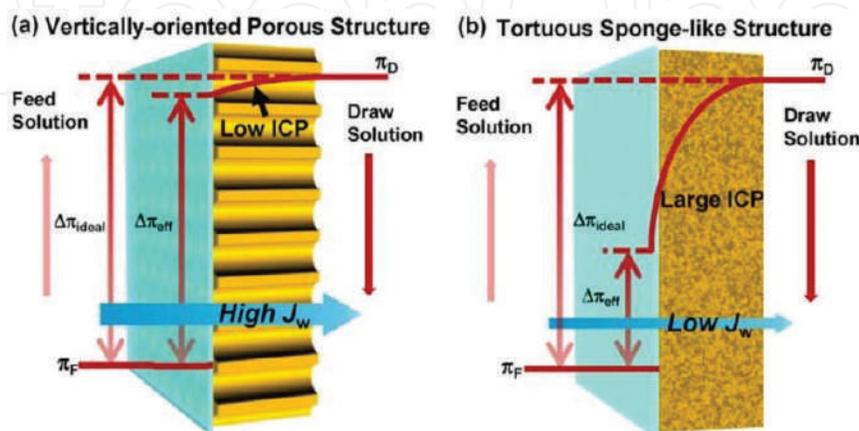
The selective layer exhibits an unprecedented water flux up to 93.6 L/m<sup>2</sup>.h (LMH) (**Figure 12**) when driven by a 2 M NaCl as DS. This performance is evidenced by the FO membranes reported in the literature and commercially available. The authors suggested according to the results that substrates with vertically oriented porous structure are ideal supports for



**Figure 12.** (a) Water flux and (b) reverse salt flux of the FO membranes in a process mode of the selective layer facing the FS (VOPS: vertically oriented porous substrates) [77].

developing FO membranes with lower ICP and ultra-high water flux. Proposed lower ICP in vertically oriented porous layer compared with a tortuous sponge-like structure in FO membranes by means of salt concentration profile are illustrated in **Figure 13** [77].

In a more recent study, Kwon et al. [78] produced a highly permeable and mechanically resistant TFC-FO membrane with a new support layer which has been already commercialized porous polyethylene (PE) membrane as the lithium ion battery separator. The very open and interconnected pore structure of the PE support, when combined with the thickness (~8 μm), is useful for alleviating the ICP, thus increasing the FO water flux. The use of a suitable plasma treatment and a surfactant in the PE support resulted in a stable formation of a PA permselective layer on the support by IP process. The prepared PE supported TFC (PE-TFC) membrane exhibited high water flux and low reverse salt flux performance due to its significantly low structural parameter. The performance values obtained in this study are also compared with other flux values in the literature (**Table 3**). The PE-TFC membrane has superior mechanical properties compared



**Figure 13.** The salt concentration profiles of (a) VOPS-TFC and (b) PI-TFC FO membranes.  $\pi_D$  and  $\pi_F$  denote the osmotic pressure of draw solution and FS, respectively.  $\Delta\pi_{ideal}$  indicates the osmotic pressure difference between the bulk feed and the bulk draw solution.  $\Delta\pi_{eff}$  means the effective osmotic pressure driving force due to the presence of ICP effect [77].

Membranes	S ( $\mu\text{m}$ )	$J_w$ (LMH)	$J_s/J_w$ (g/L) (FO/PRO)	Refs.
PE-TFC	161	41.9/45.1	0.50/0.49	[78]
PES hollow fiber-TFC	219	26.5/37.6	0.17/0.14	[64]
PAN flat sheet-TFC	350	28.8/36.3	0.10/0.13	[79]
Cellulose ester flat sheet-TFC	32	56.9/89.5	0.14/0.12	[52]
PESU-co-sPPSU flat sheet-TFC	324	20.0/25.0	0.12/0.13	[80]
PTA-co-POD flat sheet-TFC	236	37.5/78.4	0.15/0.15	[81]
PSf/SPPO (50 wt% SPEK) flat sheet-TFC	381	16.0/32.0	0.28/0.19	[82]
PSU/SPEK (50 wt% SPEK) flat sheet-TFC	107	23.0/34.0	0.18/0.20	[65]
PES/SPES (50 wt% SPES) flat sheet-TFC	245	25.2/33.7	0.28/0.26	[83]
Polydopamine-coated PAI flat sheet-TFC	456	14.0/48.0	0.44/0.17	[84]
PSf-silica NPs flat sheet-TFC	216	31.0/60.5	0.24/0.26	[85]
PSf-zeolite flat sheet-TFC	340	33.0/65	0.55/0.47	[86]
PSf-LDH flat sheet-TFC	148	18.1/34.6	0.45/0.36	[87]
PES nanofiber-TFC	106	46.0/50.0	—	[72]
Nylon 6,6 nanofiber-TFC	—	21.0/27.0	0.24/0.44	[88]
PVDF nanofiber-TFC	193	22.0/31.0	0.17/0.43	[89]

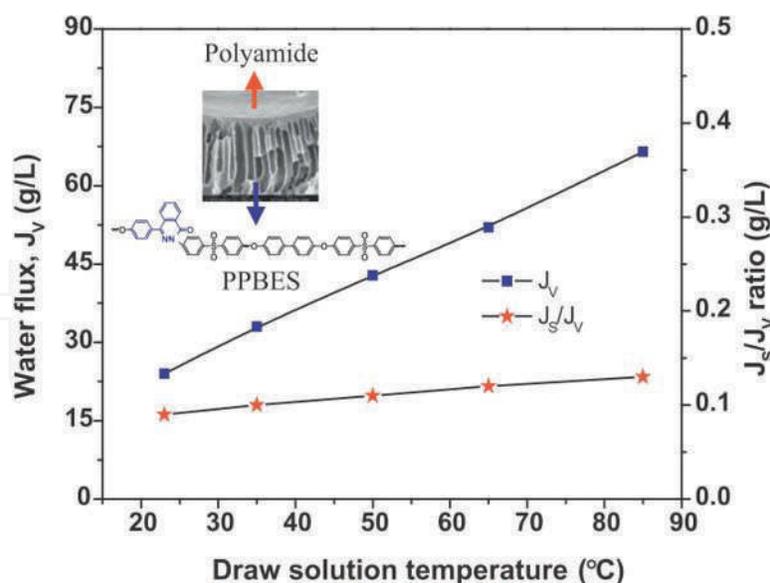
**Table 3.** Comparison of the FO performance of RO-grade membranes (FS: DI water, DS: 1.0 M NaCl) [78].

to the much thicker commercial FO membrane due to the exceptionally high mechanical integrity of the PE support. The proposed strategy offers a new material platform for FO membranes with strong commercial potential and excellent performance and durability.

In another recent study on the support layer, Zhang et al. [90] manufactured hollow fiber FO membranes with improved thermal stability using IP process on the lumen side of the co-poly (phthalazinone biphenyl ether sulfone) (PPBES) substrate. The increase in water flow in the PPBES substrate also increased the flow in the FO membrane. IP preparation parameters such as solvent, monomer concentrations, reaction time and curing conditions have been shown to seriously affect the development of composite FO membrane properties. The water flux of composite FO membranes increased from 24.0 to 66.5 LMH without a significant change in salt flux/water flux ( $J_s/J_w$ ) ratio when the draw solution temperature was raised from 23 to 85°C (**Figure 14**).

### 3.2. Active layer properties and manufacturing techniques

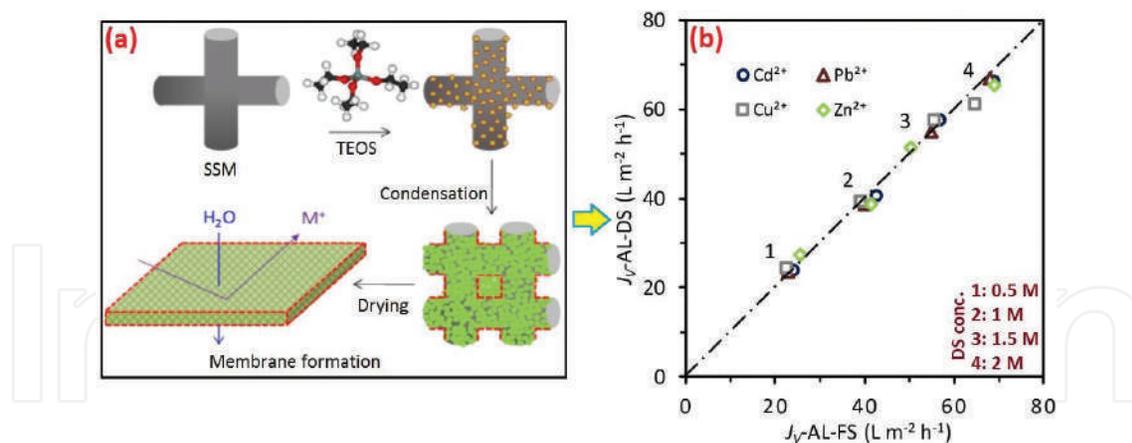
Li et al. [60] in their comprehensive review stated some recent applications on manufacturing or modifying active layer of FO membranes. Accordingly, the preparation of PA TF FO membranes is also similar to the preparation of TFC RO membranes. It is necessary to optimize the parameters such as the reaction time and the air drying duration and compositions of the monomers. Klaysom et al. [91] noted that both the surfactant additive and the drying of excess amine solution prior to the reaction are two critical parameters in PAN support preparation to control membrane properties. The addition of SDS increases polymerization and helps to form



**Figure 14.** Water flux changing of thermally-stable FO membrane with draw solution temperature [90].

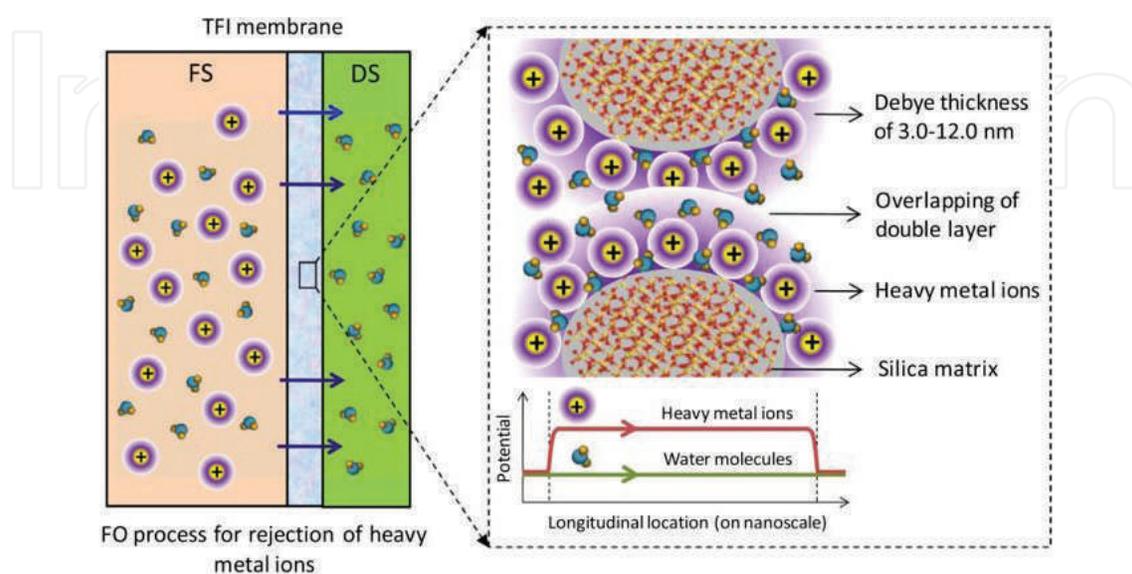
a uniform and highly cross-linked PA film. Thus, the rate of salt uptake in this study increased from 57% to over 95%, and the decrease in permeability did not occur. On the other hand, the removal of the excess amine solution before interacting with TMC resulted in the formation of a less rough membrane with improved salt rejection [91]. Due to the ionic interaction between cetyltrimethylammonium chloride (CTAC) and *m*-phenylenediamine (MPD) in the aqueous solution, CTAC may alter the reaction of the monomers of the presence and polymer molecular aggregation. Increasing the CTAC content improves the formation of the linear PA structure and microcrystalline structure of the active layer, but consequently the water flux of the PA TFC hollow fiber membrane with PES support layer is reduced, despite the high reverse salt selectivity [92]. Thermal annealing after SDS/glycerol treatment on TFC FO membranes facilitates the removal of residual unreacted monomers from the surface of the active layer, increasing the free volume size/fractional free volume ratio and reducing the total membrane thickness; so that the water flux can be improved without losing the rejection performance of the membrane [52]. Another major problem encountered in FO processes is membrane fouling, although it is less severe and reversible compared to RO processes. The structures of the support layers also significantly affect the active layer properties and hence the fouling characteristics of the TFC FO membranes. Surfaces with high roughness and large leaf-like structures are more prone to foulant accumulation and exhibit a dramatic decline in flux through these membranes, making it more difficult to improve the flux by physical cleaning of the membrane [93]. When the TFC FO membrane surface is modified, for example by covalent attachment of PEG, the tendency of surface contamination is significantly reduced due to surface barriers that adsorb pollutants [74]. On the other hand, attachment of the functionalized silica on TFC membrane via covalent amide bonds between amine groups of functionalized nanoparticles and the carboxyl groups of the TFC surface improve the fouling resistance and reduce the BSA or alginate adhesion. This is explained by the presence of the tightly bound hydration layer and the reduction of the charged carboxyl groups on the TFC membrane surface [94].

A more recent research in the literature has produced nanoporous thin-film inorganic (TFI) FO membranes with a tetraethylorthosilicate-driven sol-gel process (**Figure 15(a)**). The produced



**Figure 15.** (a) Schematic diagram of TFI membrane formation (SSM: Stainless steel mesh, TEOS: Tetraethylorthosilicate), (b) FO water flux obtained under AL-FS and AL-DS mode at initial heavy metal concentration of 200 mg/L and pH  $4.5 \pm 0.5$  for TFI membrane (adapted from You et al. [95]).

membrane was used for the removal of four typical ionic divalent heavy metals. In laboratory scale FO process, 69 LMH flux was obtained by using four heavy metal containing FS at pH 4.5 and 2 M NaCl as DS (**Figure 15(b)**). An average of 94% metal removal from the 200 mg/L FS solution was obtained. Since the hydrated ion diameters of the metals are smaller than the membrane pore size, the charge-interaction should be responsible for heavy metal rejection. Based on the classical Debye-Hückel theory and the Gouy-Chapman model, You et al. [95] have shown the importance of double-layer overlap in the membrane pore induced by electrostatic interaction between heavy metal ions and silica-made pore walls. Thus, the selectivity of the TFI membrane depends primarily on the function of the membrane pore size, the surface potential of the membrane pore wall, and Debye length (**Figure 16**). This study not only confirms the feasibility of the TFI membrane in the treatment of acidic heavy metal wastewater without pH adjustment, but it also suggests a simple theoretical scheme for better understanding and design of the charged membrane for FO applications.

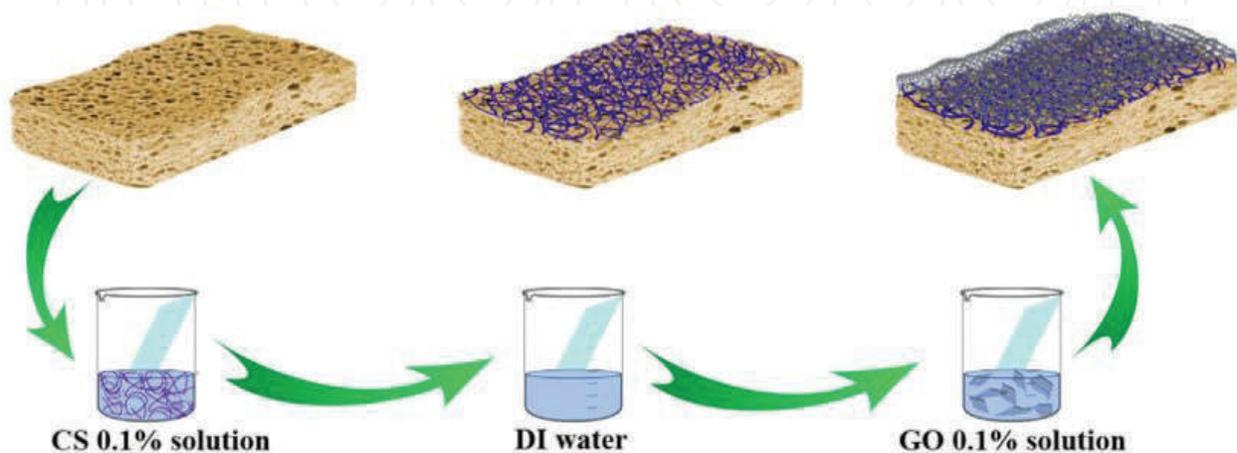


**Figure 16.** Schematic illustration of proposed mechanisms for rejection of heavy metal ions in FO process based on charge exclusion effect within the pores of TFI membrane [95].

Salehi et al. [96] in their work, fabricated a new and highly efficient FO membrane by using electrostatic interaction on a porous support layer employing layer-by-layer (LbL) assembly of positive chitosan (CS) and negative GO nanosheets. The support layer was prepared by mixing the hydrophilic sulfonated polyethersulfone (sPES) and PES using the wet phase inversion process (**Figure 17**).

Various characterization techniques have been used to confirm that the LbL membrane has been successfully fabricated. The number of layers formed in the SPES-PES support layer was easily adjusted by repeating the CS and GO deposition cycles. A TFC membrane with the same SPES-PES support layer and PA active layer was also prepared to compare membrane performances. Water permeability and salt rejection of the fabricated membranes were obtained with two types of DS (including  $\text{Na}_2\text{SO}_4$  and sucrose) for two different membrane orientations. The results showed that the membrane coated by a CS/GO double layer had a flow rate of 2–4 orders of magnitude as much as the TFC. By increasing the number of CS/GO double layers, the selectivity of the LbL membrane was improved. The newly fabricated LbL membrane showed better fouling resistance than the TFC in the FS containing 200 ppm sodium alginate as the foulant model (**Figure 18**).

Xu et al. [60] reported that generally, the flux obtained in the active layer facing draw solution (AL-DS) (PRO) configuration is higher than in the active layer facing feed solution (AL-FS) (FO) mode, but more fouling may occur in the PRO mode if the FS containing scalants/foulants is easily transported to the porous support layer. Two active layered hollow fiber membranes, one at the top of the high porosity support layer and one at the bottom, have been proposed by Wang's group [97] so that scaling or fouling can be controlled without reducing water flux in the AL-DS mode. The hollow fiber membranes with RO and NF-like scales fabricated on a PAI support were subjected to high water flux and reverse salt flux values (41.3 and 5.2 LMH) using 2 M NaCl DS and DI as FS in AL-DS mode after IP reaction and polyethyleneimine (PEI) modification. In addition, the presence of the NF-like layer on the support layer can greatly increase the resistance to scaling in the AL-DS mode. A double-skinned hollow fiber membrane with  $\text{CaHPO}_4$  scaling with a 2-hour backwash recovers 96% of the water flux while a hollow fiber membrane with a single RO selective layer has a recovery of 78% [97]. Recent studies have used polyelectrolyte LbL to form an NF-like skin in the support layer studies conducted without chemical modification, while PA-RO-like layers have also been formed. Since the resulting NF-like skin does not directly contact



**Figure 17.** Schematic illustration of CS/GO LbL assembly procedure [96].

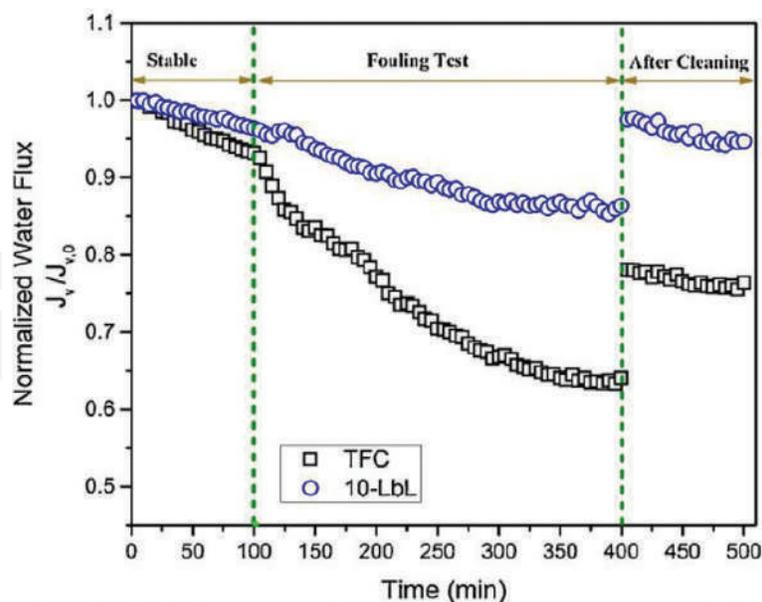


Figure 18. Fouling behavior and flux recovery of the TFC and 10-LbL membranes [96].

the FS and support layer, it prevents the transport of pollutants such as humic acid, dextran and lysozyme and thus the pore clogging. As a result, for a double-skinned hollow fiber membrane, the decrease in water flux was less than 30% during 4 hours operation, whereas for RO layer hollow fiber membranes, this reduction was 30–40% when 200 ppm foulant was used [98].

## 4. Conclusions

The active layer of an ideal FO membrane must be very thin and dense to achieve high salt retention. In order for the membrane to be able to be operated for a long time and the internal concentration polarization to be low, the support layer should be thin, hydrophilic, porous and exhibit mechanical strength as possible. The hydrophilicity must be high so that high flux and low fouling can be achieved. According to the current studies, utilizing novel nanomaterials, substrates and layer-by-layer assumptions in manufacturing of FO membrane undoubtedly enhance the water flux, rejection of the pollutants and minimize the membrane fouling but using synthetic wastewater -generally- containing one model foulant or DI water as feed solution makes it difficult to predict how FO membranes will act in real wastewaters or harsh environmental conditions. Therefore working with complex foulants and real wastewaters to better understanding of membrane behaviors, using modeling tools for fouling prediction and new cleaning strategies are essential to mitigate intrinsic challenges of the FO membranes.

In on-going researches, the developed new support layers appears continue to increase water flux slightly, however, lower water flux remains as a main challenge of the process when compared the conventional membrane systems. It is also a fact that the diffusion provided by draw solution in the process is not effective alone to increase product water volume, therefore, some promotive factors such as rehabilitated hydrodynamic behaviors or simultaneous filtration could be provided together with diffusion phenomena in further researches.

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## Forward Osmosis Membranes – A Review: Part II

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Ebubekir Yüksel and İsmail Koyuncu

Additional information is available at the end of the chapter

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### Abstract

Forward osmosis (FO) is a technical term describing the natural phenomenon of osmosis: the transport of water molecules across a semipermeable membrane by osmotic pressure from a feed solution (FS) to a draw solution (DS). The diluted DS is then reconcentrated to recycle the draw solutes as well as to produce purified water. As the driving force is only the osmotic pressure difference between two solutions, meaning that there is no need to apply an external energy, this results in low fouling propensity of membrane and minimization of irreversible cake forming, which are the main problems controverted by membrane applications, especially in biological treatment systems (e.g., FO membrane bioreactor (FO-MBR)). The purpose of the book chapter is to bring an overview on the FO membrane manufacturing, characterizing and application area at laboratory or full scales. This book chapter is published in two parts. In the second part, which appears here, characterization of mass transport in FO membranes, fouling mechanisms and foulants on FO membranes in naturally asymmetric structure and application areas of FO membranes in the literature are mentioned. Cutting-edge technologies on FO studies are comprehensively reviewed and following major and minor titles are stated truly on the new technologies.

**Keywords:** forward osmosis, characterization, structural parameter, membrane fouling, concentration polarization, water/wastewater treatment, desalination, hybrid processes, membrane bioreactor

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### 1. Introduction

FO membranes are preferred over the last few years due to the high rejection of a wide range of contaminants and the lack of hydraulic pressure, resulting in less irreversible fouling on

the membrane surface compared to pressure-driven membranes. However, due to the asymmetric structure of the FO membrane, concentration polarization (CP) becomes more important, which motivates many researchers to focus on the selection and/or development of new membrane materials for both active and support layers to decrease CP.

In this second part of the chapter, characterization of FO membranes, such as determining rejection capabilities of membrane layers by analytical approaches and experimental procedures, is thoroughly stated by considering both review and research articles in the available literature. In the following section, fouling phenomena in FO membranes are referred by considering membrane orientation, and before Conclusion, application areas of FO process are presented. Since the permeate (diluting DS) of the FO membrane is not actually a product water, this filtrate (diluted DS) needs to be treated again. For this reason, the FO process needs an additional process to recover the water from the diluted draw solution. In this context, hybrid FO processes are also included in this section. Finally, the general summary of the research is evaluated and the future prospects for FO membranes and applications are introduced.

## 2. Characterization of FO membranes

Although the model development on characterization for FO membranes is described in some literature [1], more general information from some is given here. The membrane in separation process using osmotic pressure as driving force must be capable of rejecting both the FS and the DS. When there is no solute retention in membrane, the FS and DS are easily diffused from the membrane, and osmosis does not occur. All existing membranes that can be used for this purpose are asymmetric. Many of the problems in the FO process resulted from this asymmetric structure. As with all membrane processes, mass transfer boundary layers form near the selective interface. On the FO membrane, these boundary layers occur on both sides of the selective layer interface. However, in an asymmetric membrane, one of these interfaces is embedded in the support layer. Therefore, the support layer significantly reduces the mixing and prevents the mass transfer [2]. The support layers in the TFC RO membranes are relatively thick on the FO membranes and have 25–45% porosity [3]. Solutes must be transported by support layer to reach to the selective layer on which diffusion or rejection is performed. If the mass transfer in these layers is weak, the situation called ICP occurs. Similar to conventional CP, ICP reduces the osmotic driving force. In an FO membrane where there is an asymmetric support layer in which no mixing occurs, the osmotic driving forces can be severely reduced, resulting in no water flux from the membrane [4]. The severity of ICP is greatly influenced by the support layer. This structure is often referred to a metric known as the structural parameter,  $S$

$$S = \frac{t\tau}{\varepsilon} \quad (1)$$

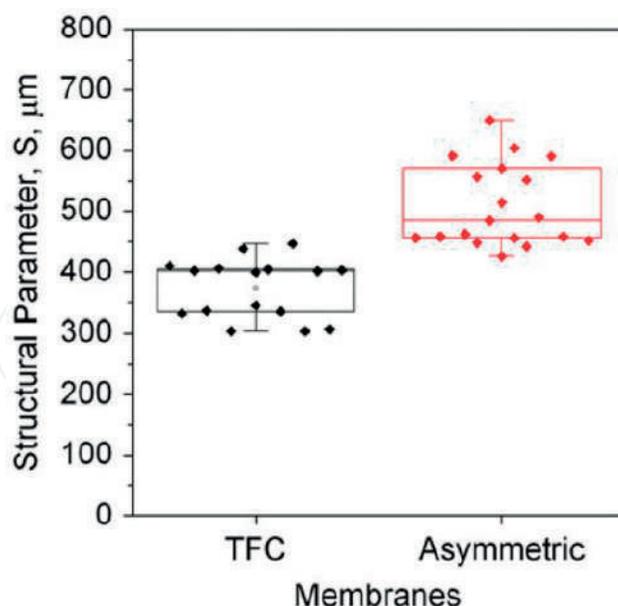
where  $t$  is the thickness,  $\tau$  is the tortuosity, and  $\varepsilon$  is the porosity of the support layer. In the FO process, membranes with lower  $S$  values are preferred to reduce ICP severity. To this end, a

number of studies have been conducted on the production and modification of new FO membranes with low  $S$  values since 1990. Tiraferri et al. [5] conducted studies on the effects of solvent quality, dope polymer concentration, backing layer wetting, and casting blade gate on support layer production on one of the first TFC membranes designed for the FO membrane. The pore morphology of the support layer was characterized with the aid of cross-sectional SEM images and reported that the optimum FO membrane must be formed from a mixed structured backing layer and that the upper part of the thin sponge-like layer should be placed on high porosity macrovoids. Shi [6] investigated UF-type phase inversion cast supports for hollow fiber FO membranes and reported that substrates with 300 kDa (molecular weight cut-off (MWCO)) should be preferred to obtain a “good” semipermeable skin.

It has also been claimed that, considering the suitability of the substrate for IP, taking into account the MWCO parameter is more appropriate than the mean pore size. It is estimated that membrane thickness is more important than porosity and tortuosity in recent studies with nanofiber membranes [7]. Moreover, the support layer pore diameter, which is thought to be very effective only in the formation of the selective layer, has also been shown to influence ICP [8]. The influence of the support layer structure on transport is typically expressed using the structural parameter concept. To calculate  $S$ , the membrane thickness (can be measured by SEM and relatively easily), porosity, and tortuosity should be measured independently. However, it is quite difficult to measure these last two, especially tortuosity, accurately and reliably. The reason for this is that the characterization of the pore structure of soft materials is an area where work is still developing and there is no standardization for the comprehensive and accurate characterization of 3D structures. Hence, researchers on FO use and develop numerical models more commonly than calculating  $S$  parameters with Eq. (1).

Experimental measurements are used when the  $S$  parameter is calculated, and therefore, the experimental conditions as a factor are emerging from the structural properties of the membrane. This means that changes in experimental conditions will directly affect the estimated  $S$  value. Therefore, no significant comparison can be made between these support layers unless the same experimental conditions are used to test different membranes. In a study by Cath et al., this limitation of the semiempirical method is clearly emphasized [9]. In this study, researchers from 7 different laboratory groups tested 2 different membranes from the same production line under the same experimental conditions but on different systems. One was an HTI-CTA membrane commercially available from HTI, and the other was a TFC membrane from Oasys Water. Significant deviations could be observed between the effective  $S$  values obtained by different groups as shown in **Figure 1**. Therefore, researchers report that the experimental conditions are the main factors in the calculation of the effective  $S$  parameter in semiempirical calculation method [1].

More recently, a simple characterization method based on a combination of a single FO test and a statistical approach has been developed to avoid pressure RO testing, which can damage the FO membrane or misread membrane properties in the characterization of FO membranes [10]. In this single test, the membrane is operated in AL-FS mode to measure water and reverse salt flux using deionized water (DI) as feed and NaCl as the DS. The statistical approach uses



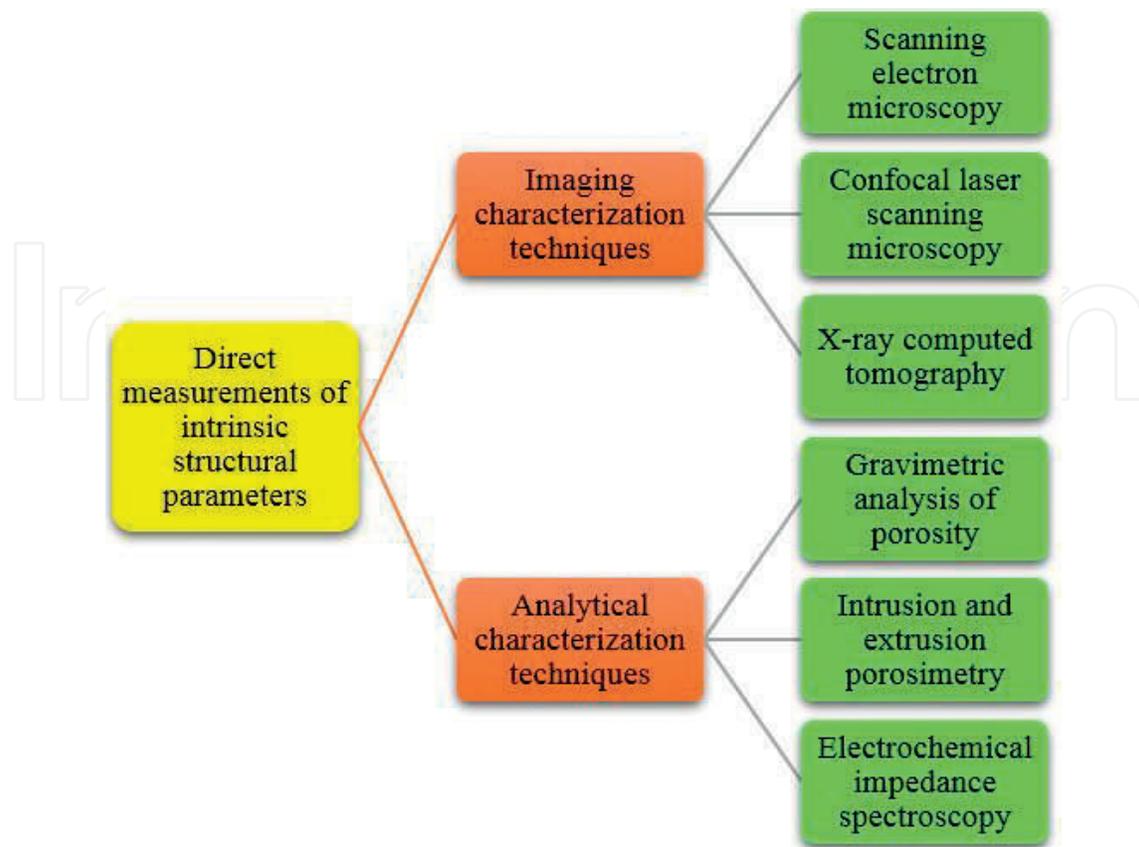
**Figure 1.** Structural parameters of TFC and asymmetric FO membranes [9].

both ICP and external concentration polarization (ECP) models to predict  $J_w$  and  $J_s$  on the tested membrane and finds the most appropriate water and salt permeability (A and B) and salt diffusion resistance in the support layer. Verifications using various experimental results in this study and other literature have shown that this new FO membrane characterization method sets parameters (A, B, and KICP) more reliably than the conventional characterization method based on the pressure-RO experiment to estimate the experimental  $J_w$  and  $J_s$ . The consideration of ECP helps to determine more accurate FO membrane parameters (especially KICP), but it is difficult to accurately model the ECP for the FO membrane channel tested.

The evaluation of porosity and tortuosity has been carried out with traditional characterization techniques such as SEM and porosimetry as well as newer tools such as x-ray computed tomography (XCT). While none of these techniques comply with all of the difficulties listed above, some are more suitable than others according to the type of the membrane material being tested. Imaging approaches provide good visuals for evaluating the qualities of porous membranes. However, expensive and time-consuming techniques are required to obtain this information from images. It also requires usage expertise. But all of these, as well as resolution and field-of-view (FOV) limitations, are disadvantages that reduce the quantitative value of these images.

Membrane pore structure analysis can also be done without relying on the images. There are a number of analytical techniques that can examine the pore structure by means of probing. While these approaches do not reintroduce visual presentation of membranes, they can provide critical characterization information about FO membrane, including porosity and tortuosity, by using basic models.

Compared with imaging techniques, analytical techniques allow for greater comparisons between different FO membrane structures by easily analyzing a larger sample volume. However, the



**Figure 2.** Direct measurement techniques of intrinsic structural parameters (adapted from [1]).

assumptions used to derive the models calculating the porosity and tortuosity must be carefully considered before adapting to the sample being analyzed. Similarly, when analyzing data from them, the biases of different analytical techniques should be considered [1]. Direct measurement techniques of intrinsic structural parameters are presented together in **Figure 2**.

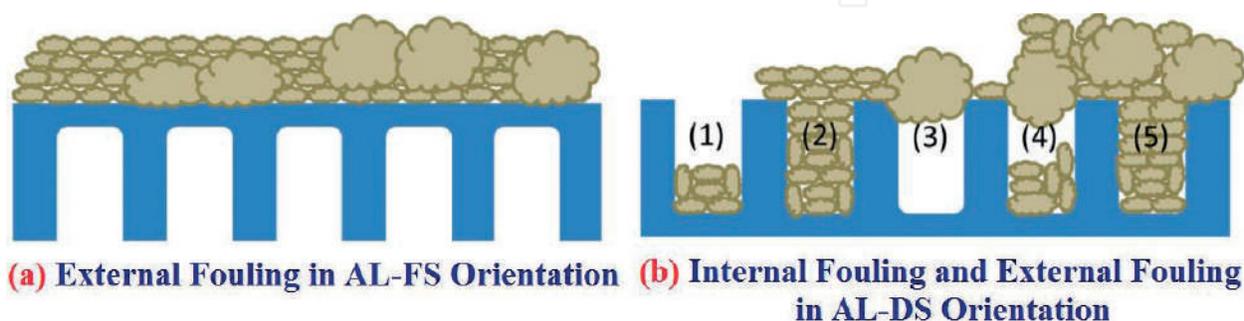
Designers of membranes for osmotic processes need to be able to better calculate the mass transfer resistance of the membrane to overcome the difficulties in nature of osmotic systems. Unless major structural parameters such as porosity and tortuosity are known, wrong areas of designs may be focused on. In order to overcome these difficulties, the above-mentioned methods for membrane characterization need to be further developed [1].

### 3. Fouling in FO membranes

Today, the greatest challenges of FO technology can be summarized into three main classes: the difficulty of developing a correct and an effective FO membrane, the lack of recyclable and economical DS, and the limited availability of information on membrane fouling [11]. Although fouling of FO membranes is more reversible than RO membranes, removal of contaminants may become more difficult when the feed stream in the FO membrane contacts the support layer [12, 13].

She et al. [14] investigated the membrane fouling in osmotically driven membrane processes and concluded that fouling in pressure-driven membranes can occur at different locations of the membrane [15, 16]. As shown in **Figure 3(a)**, the foulants in the FS are transported to the active layer surface of the membrane in the AL-FS mode, resulting in a cake layer similar to fouling of the RO membranes. This type of pollution is called external pollution. Fouling occurring in the FO membrane in AL-DS mode is more complicated. **Figure 3(b)** shows possible fouling scenarios in AL-DS orientation. If the contaminant has a relatively small size and is able to enter the porous support layer by convection of the FS, it will either be adsorbed through the walls of the pores of the support layer or eventually be retained by the active layer and accumulate on the back surface of the active layer. Subsequently, the foulants entering the porous support layer will adhere to the contaminants that are adsorbed on the walls of the support layer pores or to the accumulated contaminants on the back surface of the active layer, thus leading to “pore clogging.” This form of pollution is called internal fouling (scenario (1) in **Figure 3(b)**). In severe fouling conditions, contaminants will continue to accumulate on the outer surface of the porous support layer, as well as internal pore clogging. This type of membrane fouling is referred to as combined internal and external fouling (scheme (2) in **Figure 3(b)**). If the foulants have relatively large sizes and cannot enter the porous support layer, they may only accumulate on the outer surface of the porous support layer. In this case, only external fouling occurs (scenario (3) in **Figure 3(b)**). If contaminants are present in the feedwater in different sizes, both external fouling and internal fouling may occur (scenarios (4) and (5) in **Figure 3(b)**).

According to She et al. [15], compared to internal fouling, it is easier to remove the external fouling from the membrane surface by optimizing the hydrodynamic conditions of the feed stream (such as by increasing the cross-flow rate, applying pulsed flow [17] and employing air scouring [18]). For this reason, most researchers suggest AL-FS orientation in the FO process to prevent undesired internal fouling, even though the ICP in AL-FS is more severe than in AL-DS mode [13, 19]. However, external fouling is more reversible in FO membranes, as there is no such matter as compaction of pollution due to hydraulic pressure in the RO membrane [20]. On the other hand, the internal fouling within the porous support layer functions as an unmixed layer. Internal pollution is less reversible than external pollution, as it is more difficult to control the optimization of hydrodynamic conditions [21]. Internal fouling usually occurs in PRO membranes operating in AL-DS mode [22]. Although the osmotic backwash method has been developed to clean contaminants in the support layer [21], the development



**Figure 3.** Fouling types in FO membranes (a) FO mode, (b) PRO mode (adapted from [14]).

of more effective strategies for internal pollution control will still be an important research topic in the future.

Classification and interaction of foulants in osmotic pressure-driven membrane processes [22] can be grouped into four main classes: (i) colloidal fouling by accumulation of colloidal particles on the membrane, (ii) organic fouling by deposition and adsorption of the macromolecular organic compounds on the membrane surface, (iii) inorganic scaling by precipitation or crystallization of inorganic compounds that are poorly soluble on the membrane surface, and (iv) biofouling by adhesion and accumulation of the microorganisms to the membrane surface and eventually biofilm development. The specific pollutants in the different groups are closely related to the characteristics of the feedwater. Contaminants specifically present in raw and treated wastewaters are particles, colloids, and organic macromolecules such as polysaccharides, humic substances, and proteins [23]. In addition, these substances are also commonly found in natural waters such as rivers, seawater, and ground waters [13]. Zhou et al. [23] used gas chromatography-mass spectrometry (GC-MS) to identify soluble microbial products (SMPs) containing a large portion of polysaccharides, proteins and humic substances in raw and wastewater. Recently, organic carbon detection-organic nitrogen detection (LC-OCD-OND) has become increasingly popular for the identification of these pollutants [24]. Organic contaminants deposited on the membrane can be identified by Fourier transform infrared (FTIR) spectroscopy, solid-state  $^{13}\text{C}$ -nuclear magnetic resonance (NMR) spectroscopy, and high performance size exclusion chromatography (HP-SEC) [25]. Total organic carbon (TOC) measurement and UV analysis were also performed to determine the density of organic foulant deposition on the membrane [26]. Transparent exopolymer particles (TEPs) are another important organic pollutant typically found in natural waters. TEP in the feedwater is determined by two methods: microscopic counting and colorimetric detection [27].

Silica is a major inorganic foulant and is usually present in dissolved form or as colloidal particles in sea water, brackish water, and wastewater [24]. In addition, other inorganic contaminants are dissolved salts such as calcium carbonate, calcium sulfate, and calcium phosphate [28]. These inorganic contaminants deposited on the membrane surface can be extensively characterized by scanning electron microscopy-energy dispersive X-ray diffraction (SEM-EDX) [28] and X-ray diffraction (XRD) [29]. Microorganisms are mainly found in activated sludge in membrane bioreactors (MBR) as biofoulants [28]. These microorganisms can also be found in natural waters and cause biofouling in seawater and brackish water desalination [24]. Microbial populations within the biofilm can be characterized by analysis of DNA extracted from living cells using microbiological methods such as polymerase chain reaction denaturing gradient gel electrophoresis (PCR-DGGE) and fluorescent in situ hybridization (FISH) [23].

She et al. [15] indicated that membranes in osmotic pressure-driven membrane processes are contaminated by natural or industrial waters and wastewaters, and membrane fouling involves the combination of the four fouling categories above [24]. The understanding of mixed pollution mechanisms is difficult because of the various and numerous pollutants. Many studies to understand these fouling mechanisms are generally based on the consideration of a single foulant and the use of a synthetic FS [13, 19]. Meanwhile, the number of studies on fouling of FO membranes is also increasing. In particular, osmotic MBR studies have the

potential to conduct research with more complex wastewaters [28, 30]. Working with a single model of foulant is more advantageous in terms of easier control of the selected foulant and understanding of the foulant-foulant or foulant-membrane interactions. The physicochemical properties are also important factors affecting the stability of contaminants in the FS, as well as information on the tendency to contaminate the membrane [31]. With the understanding of the fouling mechanisms in a single foulant system, future studies may focus on the study of the fouling mechanisms for mixed foulant systems, which may lead to a better understanding of the membrane fouling mechanisms.

Colloidal and organic fouling with highly complex mechanisms in FO membranes is affected by a number of physical and chemical factors, and in general, these factors can be divided into five groups: (i) operating conditions such as initial water flow, cross-flow rate, spacer features, ventilation, and temperature; (ii) feedwater characteristics such as foulant type, concentration, pH, temperature, ionic strength, and ionic composition; (iii) DS properties such as solute type and concentration; (iv) Membrane properties such as structural and surface characteristics; (iv) membrane orientation as AL-FS and AL-DS [31].

The composition of the FS is one of the most important factors affecting membrane fouling. The effect of the feedwater composition on FO membrane fouling is similar to that of pressure-based membrane processes, and recently some investigations have been conducted on this topic [13, 32]. Generally, the degree and rate of fouling are strongly dependent on the properties and concentration of pollutants in the feedwater. In addition, since the FS chemistry significantly affects the physico-chemical properties of the contaminant [22, 33], it will also play a role in foulant-foulant and foulant-membrane interactions and determine the membrane's fouling behavior.

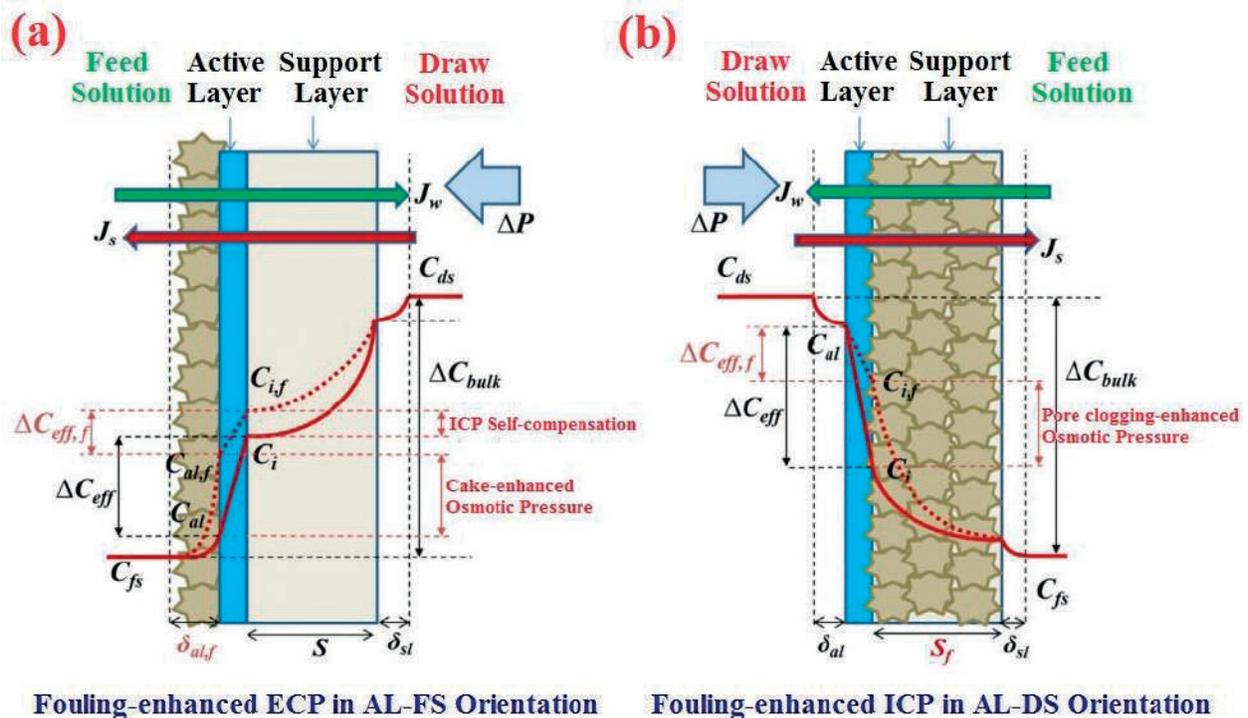
The composition and concentration of the DS, the main source of osmotic pressure in the FO process, not only affects water and salt flux but also plays a role on the membrane fouling. In general, as the DS concentration increases, the initial water flux increases and exacerbates membrane fouling. Studies in the literature have reported membrane fouling increases with increasing DS concentration [13, 19, 34]. The high hydraulic drag force caused by the high flux also leads to the accumulation of foulant on the surface of the membrane. In this context, the change in DS concentration leads mainly to changes in hydrodynamic conditions. For this reason, pollution behavior due to DS concentration can be well explained by the flux-dependent fouling mechanism in which hydrodynamic conditions play a dominant role.

Membrane material and properties may also affect membrane fouling behavior. Membranes used in osmotic processes are generally originated from a nonporous active layer formed on a porous support layer [35–37]. The intrinsic separation properties of the active layer and the structural properties of the support layer govern the transport of water and solutes, which may affect membrane fouling behavior. Membranes with superior separation properties and structural properties (i.e., more water permeability, high selectivity, and membranes with smaller structural parameters) can provide higher water flux. However, the increased hydrodynamic drag force due to increased water flow will also increase the membrane fouling potential. On the other hand, membranes with low separation and selectivity properties may increase the risk of membrane fouling as there may be more solute transfer between DS and

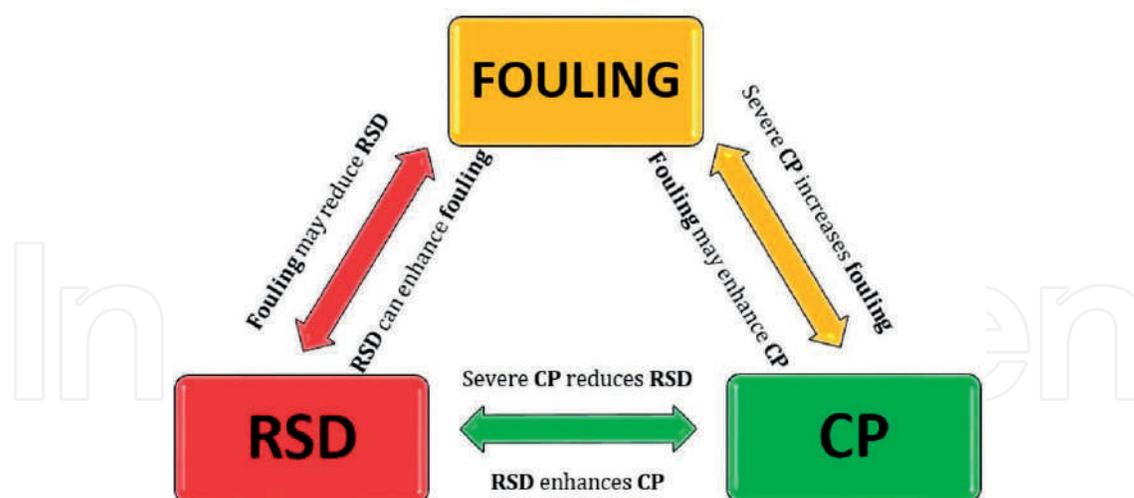
FS. When designing or selecting membranes for FO applications in the future, the separation and structural properties of the membranes should be considered not only in terms of water flow performance but also in terms of the fouling behavior [15].

Membrane fouling and CP behave differently in different orientations of membrane (AL-FS or AL-DS) in osmotic pressure-driven membranes (**Figure 4**). Therefore, fouling and CP are defined as cake-enhanced external concentration polarization (CE-ECP) in the AL-FS mode [38], while in the AL-DS mode, it is defined as pore clogging-enhanced internal concentration polarization (PCE-ICP) [19]. It is reported that the main factor that dominates water flux in osmotic pressure-driven membranes is ICP and PCE-ICP presumably plays a leading role in the flux declining. Furthermore, while CE-ECP is very effective in AL-FS mode membrane fouling, a strong ICP effect can moderate flux decline rate. On the other hand, PCE-ICP can cause much more severe flux declines. However, systematic studies are still needed to explore the effects of CE-ECP and PCE-ICP on membrane clogging in osmotic pressure-driven membranes.

As shown in **Figure 5**, membrane fouling, CP (both ICP and ECP), and RSD are closely interrelated and can be modeled using the osmotic-resistance filtration model. Factors and mechanisms affecting FO membrane fouling such as hydrodynamic conditions, feedwater composition, membrane properties, and cake-enhanced concentration polarization (CE-CP) are also applicable for NF/RO processes. Osmotic pressure is the indispensable parameter for osmotically driven membrane processes. The composition and concentration of this solution may also affect other factors by means of membrane fouling. This is the point where osmotically driven



**Figure 4.** Schematic illustration of concentration profile across the membrane due to fouling-enhanced concentration polarization (a) fouling-enhanced ECP in AL-FS orientation. (b) Fouling-enhanced ICP in AL-DS orientation (adapted from [14]).



**Figure 5.** The intrinsic interrelationship among membrane fouling: CP (concentration polarization) and RSD (reverse salt diffusion) (adapted from [14]).

membranes are separated mainly from hydraulic pressure-driven membranes in terms of the fouling mechanism. Membrane orientation (AL-FS/AL-DS or FO/PRO) is another factor affecting membrane fouling, and FO mode is more preferred as it is less prone to fouling and provides a more stable water flux. However, PRO mode can also be preferred for strong membrane stability under high pressure and fewer ICPs. However, this mode has a tendency for internal fouling, which is less reversible. Both the size exclusion mechanism and CE-CP can affect membrane fouling, which can increase or decrease the rejection of contaminants. Modification of the membrane may be a strategy to reduce the fouling of the membrane and to increase reversibility of membrane fouling, which facilitates membrane cleaning [14].

#### 4. Application areas of forward osmosis membranes

FO can be applied for the treatment of various kind of wastewaters including strong industrial effluents, i.e., from textile processes, oil and gas well fracturing waste streams, landfill leachates, nutrient-rich liquids, activated sludge, municipal wastewater, and even nuclear-origin wastewaters have been mentioned [39]. The applications of FO process can be classified as in **Figure 6**. The FO membrane rejects particles, pathogens, and emerging substances with an average porosity of 0.25–0.37 nm [40]. FO is also able to reject high levels of salt that cannot be achieved by normal treatment systems, and the total dissolved solids (TDS) from complex water can effectively be removed [41]. FO is not required for pretreatment of feedwaters (wastewaters) with complex contents. Conversely, RO and NF processes are more susceptible to fouling. Pretreatment is required to increase membrane lifetime and reduce costs [42]. FO can also be used for dewatering applications [43], useful for an efficient anaerobic digestion of wastewater, and is simpler and more environmentally friendly than classical dewatering processes [11]. High saline currents can be processed by the FO, not by the RO [44].

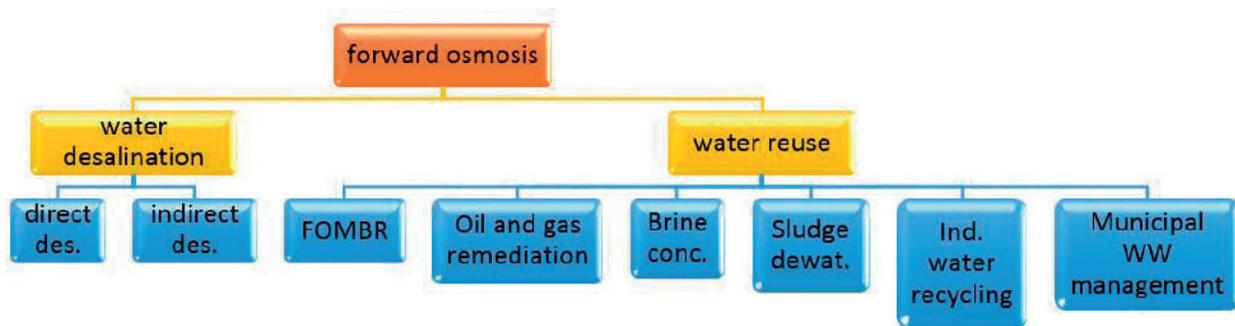


Figure 6. Forward osmosis applications (adapted from [45]).

#### 4.1. Water/wastewater treatment

According to FO literature in the last 10 years, about 7% of the studies have used complex water. However, the number of studies on wastewater is also increasing. The advantages of FO observed in these studies encourage they prefer FO instead of current technologies in future studies [45].

In municipal wastewater treatment processes, integrated FO-membrane distillation (MD) system is applied for sewer mining. In a continuous operating period, a stable water flux has been achieved at a recovery rate of up to 80% [46]. FO rejects most organic pollutants at a moderate level, whereas MD rejects almost the entire residue. Recovery of clean water from secondary wastewater was performed by FO electro dialysis (FOeED)-integrated system powered by photovoltaic energy source. This process removed total organic carbon from wastewater and produced fresh water [47]. Utilizing natural energies (osmotic pressure and solar energy), this hybrid system is a convenient process for potable water supply in isolated areas, remote areas, and islands.

MBR, which contains both activated sludge process and membrane filtration, has become one of the most widely applied technologies in wastewater treatment. The integration of the biological system with the FO membrane (FO-MBR or OMBR) can reduce energy consumption in conventional MBR. In recent years, studies on FO-MBR have been increasing [48–50]. This process not only reduces the cost of MBRs used by UF or MF but also provides fouling control through air cleaning in conventional MBR; at the same time, a more stable flux is obtained. Thus, with the help of the FO membrane in the MBR, more efficient removal efficiency is obtained with less fouling tendency without the need for hydraulic pressure [45].

FO was tested for dewatering of the nutrient-rich anaerobic digester concentration [51] in which organic compounds are rejected by FO membrane, and an RO membrane can be used to recover fresh water from a clean and diluted DS. The FO membrane was also used for activated sludge dewatering [52]. The EDTA sodium salt has been tested as DS for dewatering of activated sludge with high nutrient content. The nutrients in the sludge were successfully removed by means of FO membrane. The macromolecular DS can be posttreated with an NF process for the recovery of freshwater. Alternatively, the concentration of the RO membrane

was used as DS in Zhu's investigation and an effective sludge thickening was obtained. Thus, RO concentration is also osmotically diluted and safe disposal is possible while the volume of sludge is reduced by that study.

Another important source of pollution for wastewater treatment plants is industrial wastewater. In the US, a company has installed a pilot FO plant for the recycling of dye containing wastewater from textile and carpet mill processes [53]. In another study, the FO process was used to recover heavy metals from industrial wastewaters [54]. The effects of hydrodynamic conditions, organic pollution, temperature, and FS and DS properties on the separation efficiency were investigated. It has been reported that almost all metals such as Pb, Zn, Cu, and Cd have been removed in the study and that the FO process has the potential to be an effective and economical process for the treatment of industrial wastewater.

Linares et al. [45] expressed that, today, most FO applications for industrial wastewater treatment are devoted to the treatment and recovery of wastewater from the oil and gas (O&G) industry. In these applications, capacity for the treatment of emulsifier oil waters with FO has been stated [55]. Fresh water was recovered from wastewater by FO membrane containing up to 200,000 ppm of oil and a reasonable water flux value about 12 LMH was obtained. Many studies at the laboratory or commercial scale have been directly applied to the real wastewater of the O&G industry. Combined with RO in a closed loop, FO was used for drilling wastewater treatment from the gas exploration process [56]. The wastewater recovery capacity of the plant is 242,000 gallons of water per day, reducing the need for additional fresh water. Similar studies and applications have been performed by different companies and research groups using different membrane materials, modules, DS, and process configurations [57–59]. In these studies, it was reported that the volume of wastewater was greatly reduced, the need for fresh water was reduced, and a well-designed FO process could be a much more advantageous option than RO [60].

## 4.2. Desalination

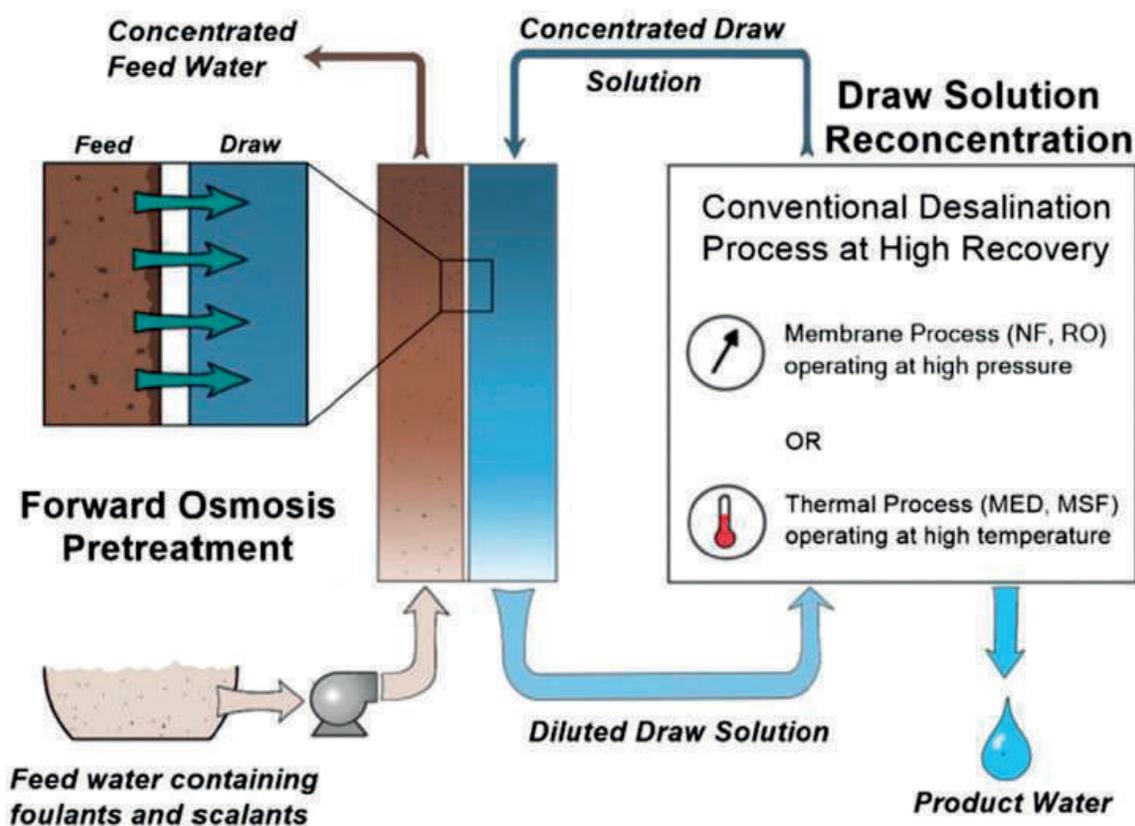
Conventional desalination technologies include membrane-based separation processes such as RO, NF, and electrodialysis and thermal desalination technologies such as multieffect distillation (MED), multistage flash (MSF), and mechanical vapor compression (MVC). Pretreatment of feedwater has critical precaution to prevent the physical equipment of conventional processes from being damaged by wastewater components and to facilitate their performance by maintaining the consistent quality of the pretreated feedwater. Today, pretreatment technologies for desalination are designed to reduce the potential for contamination of feedwater by removing natural organic matter and suspended solids. However, pretreatment technologies are typically not designed to remove dissolved solids [61]. Inorganic scaling in membrane and thermal desalination processes caused by low solubility dissolved salts in food water limits operating conditions and system performance. In MED and MSF, scaling reduces heat transfer efficiency and system recovery rates and limits operating temperatures [62–65].

Shaffer et al. [65] notified that to prevent the harmful effects of the scaling, the FO pretreatment can act to remove dissolved organic material and dissolved inorganic scalants in addition to suspended solids from the FS. When the FO process is used for pretreatment, the traditional desalination process used for recovery of the DS is only affected by NaCl solution

or an ammonia-carbon dioxide solution with negligible fouling and scaling potential of these engineered DS. The reversibility of the FO fouling shows that it can maintain the flow and performance of the FO membranes when they come in contact with raw feedwater with high fouling potential, under proper hydrodynamic conditions. A schematic view of the FO process applied for pretreatment prior to a classical membrane or thermal desalination process is presented in **Figure 7**.

The use of the FO process as pretreatment can improve the performance of conventional desalination processes by removing the small amounts of scalants present in the feedwater. The combined desalination processes can be operated at higher pressures or temperatures without the risk of scaling, resulting in higher system recovery. Testing the process modeling of an FO-RO system [66] and testing both bench-scale FO-RO [67] and FO-NF [68] systems proved the feasibility of pretreatment of the FO process. Furthermore, when FO is used instead of processes such as ion exchange and NF in the pretreatment, there is also the advantage that not only specific cations or anions but also all ions in the feedwater can be removed, in addition to the low membrane fouling tendency [45].

Linares et al. [45] notified that the direct use of FO for desalination is similar to the use of RO and NF processes conventionally used to obtain fresh water from sea water directly. This process uses seawater as FS, while nonvolatile NaCl or volatile ammonia-carbon dioxide is used as the DS [69]. However, in this process, an additional operation is required to recover the DS



**Figure 7.** Schematic of FO pretreatment for a conventional membrane desalination process [65].

from the diluted DS solution to obtain fresh water [70]. One of the most common desalination studies is the use of ammonia-carbon dioxide solution as a DS and recovering fresh water with a thermal process and regenerating the osmotic agent [2, 71]. In another study, it was reported that the total equivalent work requirement of this process was less than the conventional desalination process, such as RO, and achieved energy savings of up to 85% when expressed in terms of energy [72]. Researchers have investigated the CP in the direct desalination FO process in which ammonium bicarbonate is used as a DS and have concluded that recovery of fresh water from saline water by FO is a fairly feasible method [73]. There are also different and new DS solution searches to perform an easier and more sustainable DS regeneration in direct FO desalination studies. Generally, an ideal DS should be easy to recover and reusable with high osmotic pressure and high resolution, not toxic, easily available, and inexpensive [70]. In a study where hydrophilic nanoparticles were used as FS for DS and synthetic seawater, about 93% of salt recovery was obtained with flux and UF at around 6 LMH [74]. In a study where divalent salts such as  $\text{Na}_2\text{SO}_4$  were used as DS and brackish water as FS, 98% of the DS was rejected using NF, while 8–10 LMH flux was obtained [68]. Most DSs investigated for direct FO desalination were not commercially viable due to their high cost, limited maximum water flux they could produce, or low recovery of DS efficiencies. The world's only commercial FO facility for direct sea water treatment, was established in Al Najdah, Oman. This plant is still in operation and has reduced chemical consumption and provides longer membrane life and lower carbon footprint [75] compared to competing technologies such as traditional high-pressure RO membrane systems, saving significant operational and capital costs. These advantages have been associated with the reduction of RO membrane fouling due to the use of FO as a pretreatment step. In the direct FO desalination, similar to the RO desalination, a pretreatment process may be required. Currently, there are very few studies using natural seawater in direct FO desalination. For this reason, the fouling tendency of the FO membrane in these conditions has not been adequately investigated. However, Li et al. [24] reported that a foulant matrix containing natural organic matter and polymerized silica was formed on the membrane when natural seawater was used as feedwater.

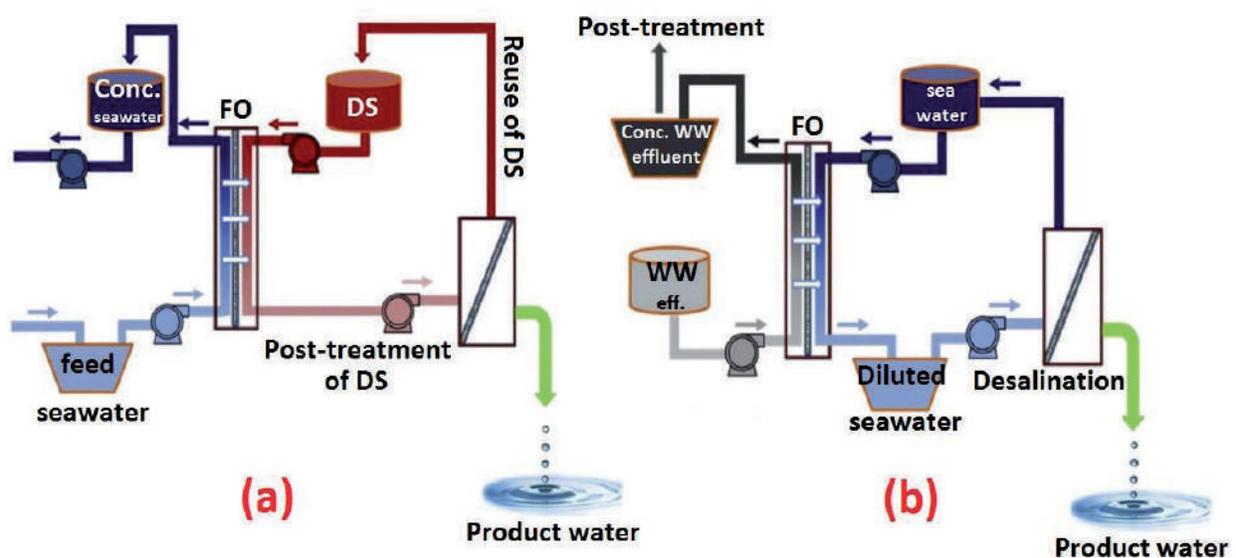
In the indirect FO desalination, there is a degraded matrix, such as wastewater or urban stormwater runoff, on the FS side, while DS is using high salinity solution [54, 76]. Potential DS in indirect FO desalination is seawater and brackish water. In addition to being free of charge DS, the main attraction is fresh water recovery through free osmotic energy from the FS, and then a partial dewatered water (diluted DS) that can be desalinated by a low-pressure RO [77]. Thus, the cost of the entire desalination process is also reduced. These studies show that FO desalination integrates fresh water treatment operations from wastewater treatment and seawater, providing a water-energy nexus for coastal cities and a promising process [54, 76].

These studies, in particular the use of primary wastewater as FS for FO, have introduced a concept of the feasibility of FO membrane, which can avoid high-cost treatment of wastewater by conventional treatment processes. For example, an anaerobic process that can be used to treat concentrated primary wastewater (concentrated FS) will provide both biogas production and reduced wastewater treatment costs [78]. Indirect desalination experiments have demonstrated the ability of FO membranes to reject waste water nutrients, especially COD and phosphate and moderately nitrogen. In addition, Linares et al. [76] could adapt the system

to the primary clarifier tank using a submerged membrane module, in partial desalination of seawater. This study also showed that FO membranes could reject up to 98% of heavy metals in wastewater. Direct and indirect layouts of desalination systems employing FO membrane are shown in **Figure 8**.

According to a fractional organic carbon analysis carried out in the fouling layer of the FO membrane, it has been reported that this fouling is mainly composed of biopolymers and protein-like substances. A similar result was observed in the FO membrane in the osmotic MBR that was used for municipal wastewater treatment [28]. When the FO system is combined with a low-pressure RO system, this hybrid process has been found to function as a double barrier against selected microcontaminants including pharmaceutically active compounds, hormones, and other organic micropollutants [79]. In practice, most of the micropollutants are rejected by FO membrane using secondary municipal wastewater as FS and sea water as DS, and removal rates were 44–95% for hydrophilic neutral compounds, 48–92% for hydrophilic neutral contaminants, and 96–99% for hydrophilic ionic microcontaminants.

In the FO process coupled with low-pressure RO, the removal of low molecular weight hydrophilic neutral micropollutants was effective (>89%) and the removal of the remaining compounds was over 99% [80]. A membrane cleaning protocol was investigated in the FO application in which municipal secondary wastewater was used as FS and sea water was employed as DS for removing of NOM-fouling through the active layer and removing of transparent exopolymeric particles from the support layer by reporting many cleaning procedures. Osmotic backwashing did not seem to help the recovery of water flux. However, when air was scoured in concentrated wastewater for 15 minutes as a cleaning technique, 89.5% flux recovery was achieved. Cleaning of the active layer with Alconox and EDTA chemistry slightly increased pollution reversibility (93.6%). The chemical cleaning of the support layer removed the reversible pollution of SL up to 94.5%. The irreversible pollution rate in these experiments was 5.5% and it was attributed to biopolymers and trace TEP that cannot be removed from the



**Figure 8.** Layout of the two FO processes for desalination (a) direct, (b) indirect (adapted from [45]).

membrane surface [18]. It has been reported that the source of irreversible contaminants on the membrane surface after chemical cleaning and at negligible level is the minimal compaction and of the nature of the FO membrane [45, 79].

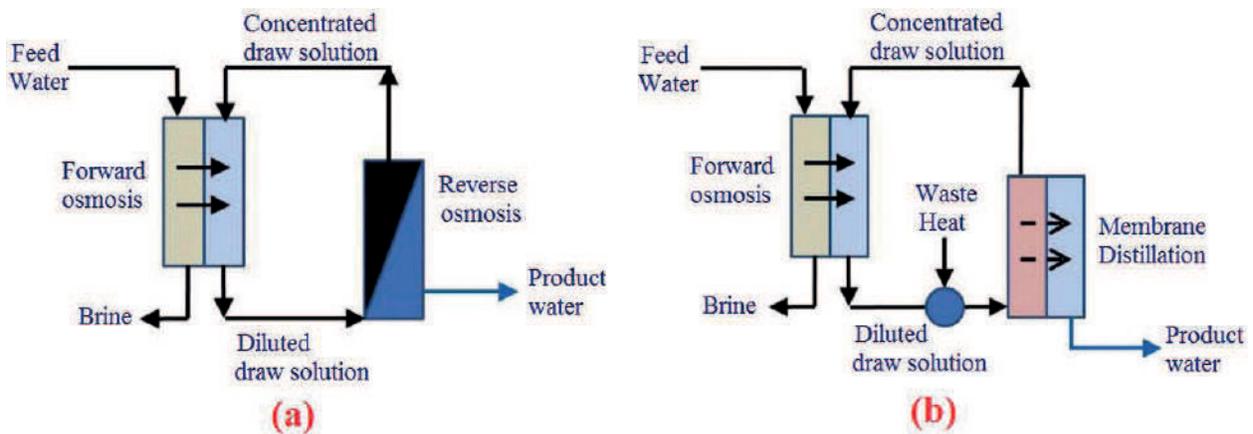
In some FO applications, the saline water is used as a major DS rather than the FS. The simplest of these applications is FO, which is used as a pretreatment for RO. In this case, seawater is used as a DS and freshwater is used as an FS for more favorable RO conditions by pressurizing and diluting sea water. Thanks to this pretreatment, the energy required for desalination of the water is greatly reduced. A similar process is pretreatment of RO water using wastewater as FS. The benefit of using such water is that the RO feedwater is diluted to more favorable operating conditions; thus, concentrated feedwater is more appropriate for effective handling. Similarly, a new procedure using ocean water to dewater an algae/nutrient solution for the production of algae biofuels is being investigated [45, 81].

In a recent analysis, McGovern and Lienhard [82] compared the specific energy consumption of a two-pass RO system with FO for desalination of seawater. At 50% recovery, for desalination of seawater containing 35,000 mg/L TDS, the two-pass RO energy consumption has been 3.0 kWh/m<sup>3</sup> including UF (for pretreatment), first- and second- pass RO. The energy consumption for the FO process with the dilution and regeneration process of DS consuming 0.10 and 3.48 kWh/m<sup>3</sup>, respectively, for the same conditions was calculated as 3.58 kWh/m<sup>3</sup>. Therefore, in order for the FO to be able to compete with the RO in terms of energy consumption, the regeneration process must be significantly more efficient than RO. However, the FO process has the advantage of having less tendency to membrane fouling compared to RO due to the lack of a hydraulic driving pressure. The FO process is also suitable for niche applications where the salinity levels of the water to be treated are higher than the salinity that can be treated by RO process [83].

### 4.3. Novel/hybrid processes

In their review on emerging desalination technologies, Subramani and Jacangelo [83] reported that the combination of the two technologies (hybrid) has shown that a hybrid technology is more effective than single use. Different hybrid configurations are being evaluated for the treatment of the hard waste waters of various industrial sources. All these industrial sectors require drinking water for various operations and applications. Emerging desalination technologies not only purify these complex wastewaters but also provide water recovery with low operating and maintenance costs and reduce the cost of electricity consumption and membrane cleaning chemicals.

Two hybrid configurations that can be used for the purification of various industrial wastewaters are shown in **Figure 9**. An FO system in **Figure 9a** is combined with an RO system for the treatment of highly contaminated wastewaters [59, 83]. Since hydraulic pressure is not present in FO, the accumulation of contaminants in the membrane is lower and the pretreatment need is eliminated. Again due to the lack of applied pressure, osmotic cleaning using a low salinity solution on the DS side will cause water transport from the DS to the FS [11]. This transport will remove loose deposits of foulants from the membrane surface and lead to more effective cleaning. The concentration of the DS is carried out using a known RO system. Because of the maximum feed pressure limit in RO, the hybrid configuration of FO and RO



**Figure 9.** Two hybrid FO applications for wastewater treatment (a) FO-RO, (b) FO-MD [83].

can only be used for the treatment of feedwater streams with low salinity. For feedwater streams with a TDS > 40,000 mg/L, DS recovery can be achieved using a gaseous NH<sub>3</sub>/CO<sub>2</sub> mixture. In this case, additional energy requirements must be taken into account in order to recover the DS using heat or other thermal methods. This configuration is particularly suitable for the refining of reflux water in the petroleum and gas industry when reuse of the water is desired. Purified water can be reused as feedwater for boilers or irrigation [83].

In **Figure 9b**, an FO system is combined with an MD system. The MD system is used for the concentration of the DS [46]. Depending on the salinity of the feed water, various DSs can be used. Since salinity is not a limiting factor for the performance of the MD system, this hybrid configuration can be used to treat wastewater with high salinity. A typical application involves flowback or processing of produced water in the oil and gas industry [84]. This hybrid configuration guarantees a minimum energy requirement when a waste heat source is available to heat the drawing solution and to reconcentrate it using MD [83].

Holloway et al. [51] suggested a hybrid FO-RO system for anaerobic digester concentration. The high energy consumption of the RO (~ 4 kWh/m<sup>3</sup>) has been a major limiting factor for the process, although water recovery has been achieved up to 75% with a high concentration of DS (70 g/L NaCl). In a further study [85], seawater was used as a DS solution in a two-stage FO process for sludge concentration to be used as fertilizer. However, high reverse salt flux and membrane fouling due to cake layer formation have been reported as serious problems of the system. Hau et al. [52] suggested a hybrid FO-NF system for a sludge dewatering application. The results showed that the FO performance was better in terms of water flux and reverse salt flux when EDTA was used as DS instead of conventional NaCl or sea water. In addition, FO has successfully rejected more than 90% of the nutrients released from the feed sludge. They also indicated that the NF recovery of EDTA sodium salts exhibiting high charged compounds performed well and had a high salt rejection of 93%. While the water flux was constant during the first hours of operation, the FO membrane was then rapidly reduced due to the increased buildup of the sludge cake layer in the concentrated feed and diluted DS.

Oasys Water Inc. has operated a pilot scale thermal-based hybrid FO system for water with high salinity (>70,000 ppm TDS), which is a product of shale gas industry [41, 60]. The results

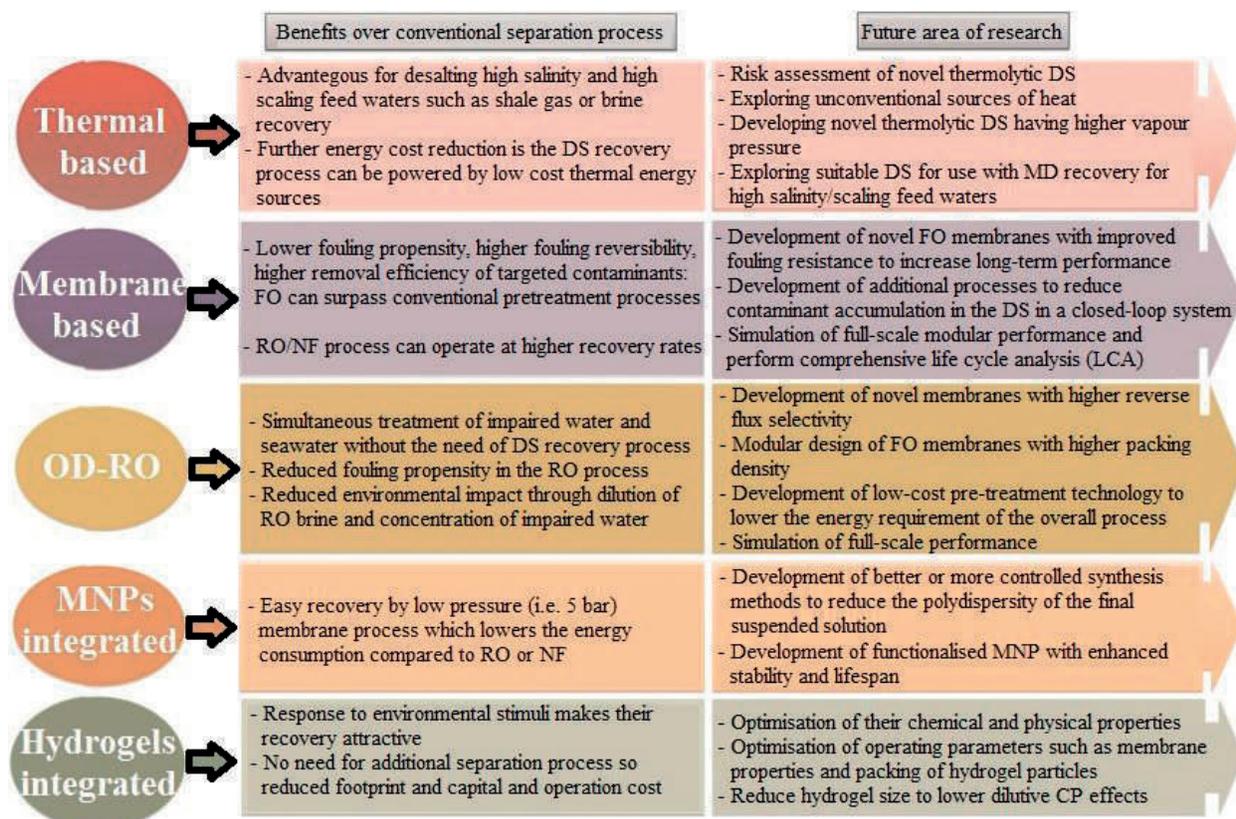


Figure 10. Summary of the benefits of current hybrid FO systems and direction for future research (adapted from [86]).

show that this hybrid system can exhibit feedwater recovery performance (60%) similar to evaporative saline concentration technologies and that the final product water meets surface water discharge criteria in terms of TDS, chlorides, barium, and strontium. However, although the RO required less specific energy when desalinating waters with lower salinity, it was found that this study was not sufficient to purify the challenging feedwater. The hybrid FO distillation system can be integrated to provide a zero-liquid discharge (ZLD) facility designed as a membrane brine concentrator (MBC). The MBC system is ideal for the oil and gas industry and provides up to 85% water recovery while discharging brine with salt concentration up to 25%. A summary of the benefits of current hybrid FO systems and direction of future research are schematized in Figure 10 [86].

## 5. Conclusions

FO process has a big potential to be an alternative solution for water/wastewater treatment and desalination purposes over conventional membrane processes. To benefit from this potential at maximum, ICP and low flux challenges should be completely solved or minimized by changing operational parameters. Changing membrane orientation (to increase water flux), utilizing various DSs (to increase osmotic pressure), and changing sludge retention time (i.e., to hinder salt accumulation in FO-MBR) are some of the basic procedures used since former FO studies.

The use of hybrid systems such as FO-RO and FO-MD even with seawater desalination and optimization energy consumption could be more feasible and better alternative than the performance exhibited by the FO process alone for wastewater recovery. However, the indispensable factor affecting the process performance is FO membrane. According to the current studies, utilizing novel nanomaterials, substrates, and layer-by-layer assumptions in manufacturing of FO membrane undoubtedly enhance the water flux and rejection of the pollutants and minimize the membrane fouling but using synthetic wastewater-generally, containing one model foulant or DI water as feed solution makes it difficult to predict how FO membranes will act in real wastewaters or harsh environmental conditions. Therefore, working with complex foulants and real wastewaters to better understand membrane behaviors and using modeling tools for fouling prediction and new cleaning strategies are essential to mitigate intrinsic challenges of the FO membranes.

In ongoing researches, the developed new support layers appear to continue increasing water flux slightly; however, lower water flux remains as a main challenge of the process when compared to the conventional membrane systems. It is also a fact that the diffusion provided by draw solution in the process is not effective alone to increase product water volume; therefore, some promotive factors such as rehabilitated hydrodynamic behaviors or simultaneous filtration could be provided together with diffusion phenomena in further researches.

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