Chapter

Shale Oil & Gas Produced Water Treatment: Opportunities and Barriers for Forward Osmosis

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Abstract

The treatment of shale oil & gas produced water is a complicated process since it contains various organic compounds and inorganic impurities. Traditional membrane processes such as reverse osmosis and nanofiltration are challenged when produced water has high salinity. Forward osmosis (FO) and membrane distillation as two emerging membrane technologies are promising for produced water treatment. This chapter will focus on reviewing FO membranes, draw solute, and hybrid processes with other membrane filtration applied to produced water treatment. The barriers to the FO processes caused by membrane fouling and reverse draw solute flux are discussed fully by comparing some FO fabrication technologies, membrane performances, and draw solute selections. The future of the FO processes for produced water treatment is by summarizing life cycle assessment and economic analyses for produced water treatment in the last decade.

Keywords: forward osmosis, produced water, desalinization, treatment technology, draw solution

1. Introduction

Shale oil & gas produced water (PW) is the wastewater generated from the oil and gas industry and is a significant portion of the industrial wastewater. During the exploration and production of oil and gas, water is injected with some chemicals to cause a hydraulic fracture, and this water is trapped underground. Then this water is brought to the ground surface and is referred to as PW. Because of the hydrofracturing conditions, the mixed water's characteristics and properties have been altered physically and chemically [1]. These characteristics vary depending on location, time, the drilling, production, treatment processes, and the geological formation in contact with the injected water. The produced water characteristics are numerous, but the most prominent are salt, oil, naturally occurring radioactive materials (NORM), organic and inorganic chemicals, metals, and various suspended and total dissolved solids [1, 2, 4]. Because of these properties, PW treatment is a very strenuous and costly process. The PW treatments can be broken down into three main stages: primary, secondary, and tertiary.

Produced water is characterized as high salinity, which implies a high concentration of total dissolved solids (TDS) (sodium, chloride, calcium, magnesium, sulfate, and other dissolvable ions or solids) [3]. Besides free, dispersed, or emulsified

oil droplets present in PW, other characteristics are calcium & magnesium causing hardness, phenols, benzene, toluene, ethylbenzene xylenes (BTEX), acids, non-volatile, volatile, and semi-volatile organics, and other chemical additives. With a wide variety of contaminants within PW, various treatment methods are required for cleaning PW instead of the deep-well injection by one or several thousand feet deep underground.

Among many processes used to cleanse PW, desalination, in which salt and other minerals are removed from the water to create freshwater, has become a forerunner. The primary method used for desalination is reverse osmosis (RO). Osmotically driven membrane processes (ODMPs), specifically forward osmosis (FO), are the inverse of pressure-driven membrane processes (PDMP), e.g., ultrafiltration, nano-filtration, reverse osmosis (RO). Pressure-driven membrane processes (PDMPs) are when high pressure is used to drive water flow through a semipermeable membrane. Inversely, ODMPs are caused by an osmotic pressure difference that pushes water flow through a semipermeable membrane from the dilute feed water (FW) to a concentrated draw solution (DS). Comparatively, ODMPs have several benefits and advantages that PDMP does not, such as low energy consumption, ease of equipment maintenance, low capital investment, high salt rejection, and high-water flux. Because of the cost and energy required in the RO systems, FO systems are being considered. FO also offers benefits such as higher productivity, rejection of various contaminants, and lower fouling tendencies.

2. Produced water

The oil and gas industry produces a lot of waste, 80% of which is a liquid waste [1]. In 2012, it was reported that the oil and gas industry in the US produced approximately 3.4 trillion liters, or 0.88 trillion gallons of PW yearly, which was 2.46 billion gallons per day [3–4]. This number is projected to continue to increase by 25% from 2006 to 2030 [1]. The oil and gas industry not only produces wastewater but uses water in several of its processes. The water produced may be discharged into the surrounding environment but must meet that region's discharge requirements. It may also be used for consumption though this would require extensive treatment.

2.1 Characteristics of produced water

There are various contaminants in PW that depend on the geological location, the geologic formation where the water is generated, and the hydrocarbon product produced. These contaminants can be organized into sections below [1–2, 4]:

- Dispersed oil
- Dissolved oil
- Dissolved minerals & metals
- Dissolved gases
- Production chemicals
- Produced solids & bacteria

Dispersed oil and dissolved organic compounds categories are frequently grouped due to their similarities. Dispersed oil refers to the dispersed oil droplets suspended in PW and can become toxic at the point of discharge [2]. These droplets are usually four to six microns but can fluctuate from two to 30 microns and are originated from the aqueous phase. Dispersed oil contributes considerably to the biological oxygen demand (BOD) and would cause an issue with the use of PW for consumption or discharging into nearby ecosystems. Dissolved oil refers to the organic compounds found in PW, and it includes phenols, BTEX, volatile hydrocarbons, polyaromatic hydrocarbons (PAHs), phenols, carboxylic acid, and other organic acids [1, 2]. These components are found in higher concentrations and are created from the platforms that produce oil rather than the plan platforms that produce gas [2]. **Table 1** illustrates the typical composition of PW samples from natural gas production.

Dissolved minerals are many inorganic compounds that naturally occur in PW. These compounds are found at high concentrations within PW and can be classified as cations and anions. Dissolved minerals can also precipitate to form solids, affecting TDS, which hinder the treatment process [2]. These inorganic compounds include heavy metals and NORMs and are typically less toxic when compared to their organic counterparts. The concentrations of various mineral ions present in shale gas PW can be seen in **Table 2**.

The main metal and minerals contributing to the high concentration of dissolved minerals are sodium, chloride, sulfate, bicarbonate, aluminum, strontium, potassium, calcium, iron, barium, and magnesium [9]. These minerals are the main contributor to the inorganic chemistry of PW, with sodium and chloride affecting its salinity and chloride, sodium, potassium, strontium, calcium, barium, magnesium, iron, sulfate, carbonate, and bicarbonate affecting its conductivity and scale-forming potential [2]. Radium-226 and radium-228 are the most abundant NORMs in PW, with barium sulfate as its frequently co-precipitated scale. Dissolved gases refer to the gases that occur naturally due to bacterial activity or chemical reactions within PW. These gases commonly are carbon dioxide, oxygen, and hydrogen sulfide [2].

Production chemicals refer to the chemicals added during oil and gas production to further aid in operation and production. These chemicals can be divided into two sections treatment chemicals and production treating chemicals. These chemicals

Parameter	Unit	Minimum value	Maximum value
pH		3.1	7.0
Conductivity	mg L^{-1}	4,200	180,000
TOC	mg L^{-1}	67	38,000
TDS	mg L^{-1}	2,600	310,000
TSS	mg L^{-1}	8	5,484
BOD_5	mg L^{-1}	75	2,870
COD	mg L^{-1}	2,600	120,000
Oil/grease	mg L^{-1}	2.3	60
Surfactants	mg L^{-1}	Not detected	285
Alkalinity	mg L^{-1}	Not detected	285

Table 1.
Composition of produced water [1–3, 5–8].

Metal/ Mineral	Minimum value (mg L^{-1})	Maximum value (mg L^{-1})
Aluminum	Not detected	83
Arsenic	0.004	151
Barium	Not detected	1740
Boron	Not detected	56
Bromide	150	1149
Cadmium	Not detected	1.21
Calcium	Not detected	51,300
Chloride	1400	190,000
Chromium	Not detected	0.03
Copper	Not detected	5
Iron	Not detected	1100
Lead	<0.2	10.2
Lithium	18.6	235
Magnesium	0.9	4300
Manganese	0.045	63
Nickel	Not detected	9.2
Potassium	149	3870
Sodium	520	120,000
Strontium	Not detected	6200
Sulfate	Not detected	47
Silver	0.047	7
Tin	Not detected	1.1
Zinc	Not detected	5

Table 2.
Minerals in produced water [2, 6–8].

can be either pure compounds or compounds that contain active ingredients dissolved in a solvent or a co-solvent [1]. Treatment chemicals are used in production treating, gas processing, and stimulation. Production treating chemicals are used in scale and corrosion inhibitors, biocides, emulsion breakers, antifoam, and water treatment chemicals. These chemicals can be seen in **Table 3**.

Solids vary from oilfield to platform and refer to the myriad of constituents produced from the formation and other operations. These solids include scale products, sand and silt, carbonates, clay, wax, corrosion products, and other suspended solids. They may consist of bacteria, which can create sulfides when the bacterial reduction of sulfate occurs [3]. However, the quantity of bacteria is usually relatively small due to various toxic chemicals within PW [1, 3]. These microorganisms found in PW are typically aerobic and Gram-positive. These solids can hinder oil production due to the oily sludge and emulsions that can clog flow lines.

2.2 Treatment of produced water

Currently, there is a myriad of treatments used for PW. The goal of these treatments are; (i) de-oiling (removing dispersed and dissolved oil), (ii) soluble

Chemical	Concentration: Oilfield		Concentration: Gas Field	
	Typical ($mg\,L^{-1}$)	Range ($mg\,L^{-1}$)	Typical ($mg\,L^{-1}$)	Range ($mg\,L^{-1}$)
Corrosion Inhibitor	4	2–10	4	2–10
Scale Inhibitor	10	4–30	—	—
Demulsifier	1	1–2	—	—
Polyelectrolyte	2	0–10	—	—
Methanol	—	—	2000	1000–15000
Glycol (DEG)	—	—	1000	500–2000

Table 3.
PW treatment chemicals [1, 9–10].

organics removal, (iii) disinfection, (iv) removal of suspended solids, (v) removal of dissolved gas (removing of light hydrocarbon gases, carbon dioxide, and hydrogen sulfide), (vi) desalination, (vii) softening, and (viii) removing NORM. The traditional treatment processes of PW can be separated by the categories:

- Physical treatment
- Chemical treatment
- Biological treatment

The main physical treatment methods for PW are physical adsorption, sand filters, cyclones, evaporation, electrodialysis (ED), dissolved air precipitation (DAP), and freeze–thaw/evaporation (FTE). The main chemical treatment methods for PW are chemical precipitation, chemical oxidation, electrochemical process, photocatalytic treatment, Fenton process, treatment with ozone, room temperature ionic liquids, and demulsifiers. The biological treatment methods for PW are the use of aerobic and anaerobic microorganisms.

Treatments outside of these three main categories are thermal and membrane filtration technologies. Thermal technology is unique to regions where the cost of energy is low-priced. Before membrane technologies reached the forefront, thermal treatment technologies were the main methods used in desalination. The main thermal technology methods for PW are multistage flash (MSF) distillation, vapor compression distillation (VCD), and multi-effect distillation (MED) [11]. Another thermal technology method is a hybrid of multi-effect distillation and vapor compression distillation (MED-VCD hybrid).

Membrane treatment technology has become a promising method for PW desalination [12]. The main membrane filtration methods for PW are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes. These can either be pressure or vacuum-driven systems with either a crossflow filtration or dead-end filtration system [1]. These membranes are microporous films made of synthetic organic or inorganic materials with various pore sizes [1–3]. The advantages of membrane filtration technology are sludge reduction, high quality of permeate, less space required, easiness of operation, minimal impact on permeate quality with variation in feed water quality, little to no chemicals needed, the possibility for recycling waste streams, the potential for having an automated plant, moderate capital costs, ability to be combined easily with other separation processes, low energy consumption, and continuous separation [2].

Among different types of membranes, MF membranes have the largest pores size from 0.1 to 5 μm and remove suspended solids and turbidity. The pore size of UF membranes range from 0.01 and 0.1 μm and these membranes remove color, odor, viruses, and colloidal organic matter. It is one of the most effective treatment methods for the removal of oil from PW. Both MF and UF membranes are made of polymeric and inorganic materials. Comparatively, NF and RO membranes are made of polymeric materials when compared to the more porous UF and MF membranes. The pore size of NF membranes range from 0.5 to 1 nm and these membranes remove multivalent ions and specifically charged or polar molecules. Additionally, RO membranes remove low molecular weight components.

3. Forward osmosis membranes

Forward osmosis is a membrane filtration process that has drawn interest in the PW treatment. The FO method is mainly used for desalination due to its ability to remove salt, minerals, and other compounds from water efficiently [13, 14]. This is because the process of FO is based on a phenomenon that occurs naturally. This phenomenon is when two solutions of varying concentrations are separated by a semipermeable membrane, and the solution with the lower concentration will pass through the membrane to the solution with the higher concentration [15]. The FO process relies on the osmotic pressure difference that causes the aforementioned phenomenon to complete the desalination task without the need of an external pressure source, unlike RO. The FO process may be used standalone for desalination or in an enhanced RO pretreatment process [13]. In FO, there are two solutions, a draw solution (DS), with high salinity and osmotic pressure, and a feed solution (FS), with low salinity and osmotic pressure, are separated by a semipermeable membrane and use the natural osmotic pressure difference to move the water from FS to DS until equilibrium is reached [16, 17]. Feed solution will increase in concentration as DS dilutes.

In general, FO membranes are asymmetric and consist of two layers, an active layer and a support layer, as shown in **Figure 1**. The most main commercially available of the various membranes used in FO are cellulose acetate (CA), cellulose triacetate (CTA), and thin-film composite (TFC) membranes [14, 18].

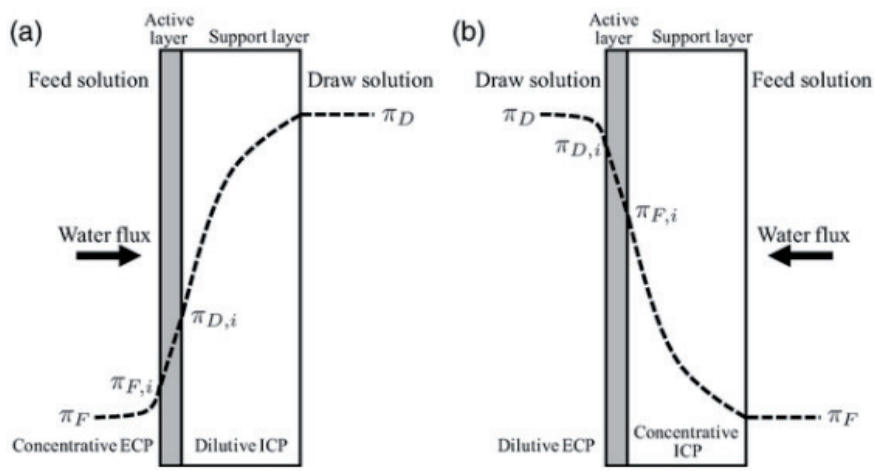

Figure 1.
Principle of forward osmosis (FO). (a) AL-FS mode, and (b) AL-DS mode [20].

Its primary purpose of the support layer is to provide mechanical support to the active layer. One imperative aspect that the support layer directly affects is internal concertation polarization (ICP). The IPC phenomenon, as to be discussed further later, is when the osmotic driving force is impaired, in this case, due to dissonance between both the active and support layer [19]. When this occurs, it frequently causes the water flux to decrease. This disconnect between the two layers is when there is a resistance in the transfer of masses from one layer to the other.

Initially, FO membranes were produced using CA, CTA, polysulfone (PS), or polyethersulfone (PES), and polybenzimidazole (PBI). And in later years, TFC membranes were used on polysulfone, sulfonated, cellulose acetate propionate (CAP), and non-woven electrospun nanofibrous substrates [16, 20–22]. The phenomenon that is osmosis was first observed in 1748 by Nollet, but no process was made towards the synthesis of these membranes until years later [23–24]. CA membranes were first hypothesized in 1957, and a case study sheds light on the possibilities this semipermeable membrane contains due to its behavior in saline water [16]. The first synthetic membrane was created in 1963 by Loeb and Sourirajan [16, 23]. This membrane was an asymmetric RO membrane, fabricated from CA, acetone, and aqueous magnesium perchlorate, with high flux and high salt rejection [16, 23]. Following this, in 1965, Batchelder utilized natural cellulose as the foundation for a semipermeable FO membrane [24].

Further employing dissolved volatile solutes, such as sulfur dioxide in seawater or fresh water, as the DS and seawater as the FS [16]. Several more studies were conducted, and in 1986 the Hydration Technology Innovation (HTI) was able to fabricate a CTA FO membrane for commercialization [26]. This membrane was used for various things, most notably as a water filtration system for global military forces and humanitarian disaster relief organizations especially [16, 23]. A new FO desalination process came to fruition in 2005, where ammonium bicarbonate was used as DS to create a high osmotic gradient across the FO membrane, and a new step was added [27]. Upon a moderate increase of temperature, ammonium bicarbonate decomposes into ammonia and carbon dioxide gases that can be isolated and reused as DS, leaving fresh PW [16]. The step added to this FO process, named indirect osmosis, is the DS regeneration, where after the DS is diluted and the concentrated DS is combined with the diluted DS [24, 26].

Over the years, more FO membranes were fabricated, tested, and made available for commercial use. CA and CTA membranes are some of the original materials used for FO applications. These membranes are still created and commercially sold today. They are not sufficient for wastewater treatment due to their operational efficiency parameters. These commercialized CA and CTA membranes provide low permeability, low salt rejection, poor resistance to biological species, and limited chemical stability [28]. PS, or PES, is another material use to fabricate FO membranes. The operation parameters for PS FO membranes are hindered because of the lack of durability and filtering capabilities. There is currently research on how to remedy these concerns using nanomaterials, such as carbon nanotubes and graphene oxide, to strengthen the membrane and increase its durability. Another important material for FO membranes is PBI. The PBI FO membranes are highly hydrophobic, and this affects their ability to efficiently be applied to water purification. Membranes that are more efficient and are more favored are polyamide fabricated. These membranes are fabricated using TFC and interfacial polymerization (IP) methods. These membranes are preferred because of their ability and because their operational efficiency parameters are much broader. They can be used under much more strenuous conditions and constraints.

3.1 Advantages of FO membranes

The utilization of FO for desalination offers a myriad of benefits when compared with pressure-driven membrane processes. One of the main benefits is the ability of FO membranes for low energy consumption due to not requiring applied hydraulic pressure. Thus, in turn, reducing the cost, especially if the DS is economically and technically viable [15]. However, this factor is contentious due to the energy required to regenerate the DS. The amount of energy utilized is comparable to the energy used in RO processes. However, this cost can be reduced by selecting a low-cost energy source for the FO process. Additionally, FO processes can be modified and integrated with other processes to reduce energy costs. Hybrid FO processes will be discussed further in Section 4.3.

Another benefit of FO that is tied to not requiring hydraulic pressure is a reduction of fouling. Recent studies have demonstrated that a lack of applied hydraulic pressure and optimization of the system's hydrodynamics can lead to less fouling and the higher reversibility of fouling [15, 29–32]. Due to the low flux that can be obtained in FO, there is more opportunity for low fouling [28, 32, 33]. Because of this, a more extensive range of pollutants can be removed from FS. The reduction of fouling can also be attributed to FO fouling layers being less dense than the fouling layers of its pressure-driven counterparts, also assisting in its reversibility. The FO processes are capable of treating FS with high fouling propensity due to their lower fouling tendency without lengthy pretreatment. The lower the fouling tendency of the membrane, the longer the lifetime of the membrane [35]. Membranes with lower fouling tendencies require cleaning and maintenance less frequently. Furthermore, the fouling on FO membranes can be physically cleaned instead of cleaning with chemicals. An additional benefit of FO is a phenomenon called retarded forward diffusion of solutes [36]. In this phenomenon, the reverse salt flux in FO processes impedes the pore diffusion of feed solutes, leading to the high rejection of feed solutes [29, 36]. As mentioned earlier, when a high osmotic pressure gradient is obtained across the membrane, sufficient water flux and recovery can be achieved [15, 34]. The increase in water recovery would cause a decrease in rejected FS, leading to a reduction in waste disposal. This would be especially beneficial for desalination plants, in particular plants offshore [15].

3.2 Challenges faced by FO membranes

Though there are many advantages to FO membranes, there are still several challenges that limit their applications. These challenges include:

- Concentration polarization
- Membrane fouling
- Reverse solute diffusion

These challenges are caused by membrane orientation and design, DS and FS concentrations, and operational conditions. Membrane orientation refers to the order in which the displacement of the FO membrane can be alternated. The AL-FS mode is where the active layer faces FS, and the AL-DS mode is where the active layer faces DS. Concentration polarization (CP) occurs in all forms of membrane separation processes, as shown in **Figure 2**. In FO membranes, CP occurs because of a concentration gradient between FS and DS through an asymmetric FO membrane [15, 18, 37]. Furthermore, CP can be broken into internal concentration polarization

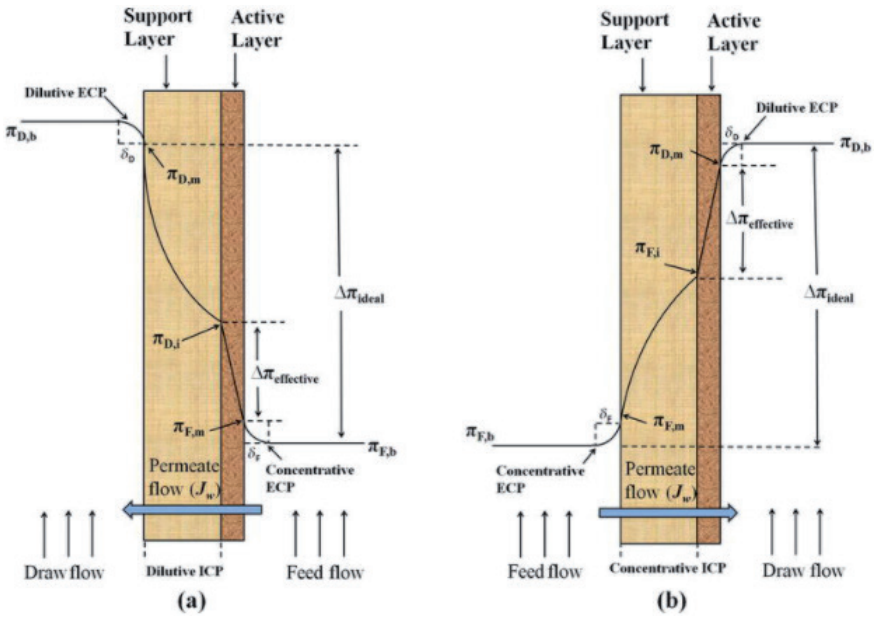

Figure 2.
Internal concentration polarization (ICP) and external concentration polarization (ECP) occurring in forward osmosis. (a) ICP and ECP in Al-FS mode, and (b) ICP and ECP in Al-DS mode [38].

(ICP) and external concentration polarization (ECP). ICP occurs within the FO membrane's support layer, and ECP occurs around the surface of the FO membrane's active layer.

The ECP phenomenon occurs around the surface of the FO membrane's active layer and is caused by a difference in the membrane surface's concentration and the concentration of the bulk solution [38]. Moreover, ECP can be defined further by two types, concentrative and dilutive. Concentrative ECP occurs in AL-FS mode and is when the FS becomes concentrated at the active layer surface as the water travel through the membrane. Dilutive EPC occurs in AL-DS mode and is when the DS is diluted on the active layer surface of the water travels through the membrane. Additionally, EPC can significantly affect the osmotic gradient, playing a pivotal role in its decrease and thus hindering water flux across the membrane [15]. Though, the amount of EPC occurring depends significantly on the DS chosen. Moreover, EPC does not have as great of an impact on the membrane's effectiveness as IPC does.

The ICP phenomenon occurs within the FO membrane's support layer and is when solutes are unable to penetrate the dense active layer, which causes the CP within the porous support layer [15, 38]. ICP can also be defined further by two types, concentrative and dilutive. Concentrative ICP occurs in AL-DS mode and is when the FS concentrates within the support layer as the water travel through the membrane. Dilutive IPC occurs in AL-FS mode and is when the DS is diluted within the support layer as the water travel through the membrane. When concentrative ICP occurs, dilutive ECP. The inverse occurs with dilutive ICP. When dilutive ICP occurs, concentrative ECP occurs.

4. Optimization of forward osmosis membranes for produced water

An ideal FO membrane consists of an active layer that high water permissibility and a support layer that minimizes CP, both internal and external, and thus enhances

mass transfer [16]. The FO membrane must also have mechanical stability, chemical resistance and less susceptible to membrane fouling. The FO membranes with these qualities have advantages over other membrane processes, such as UF, MF, NF, and RO [12]. These advantages are the capability of operating at low hydraulic pressure and an ambient temperature whilst rejecting almost all solutes and suspended solids. Because of those operating abilities, energy consumption is reduced. The cost of construction and system procedure is due to FO membranes not requiring high hydraulic pressures to overcome high osmotic pressure [39]. These capabilities translate well into harsh conditions, where there is limited access to electricity, currency, and materials. The effectiveness of the FO membrane can be described by the values of the intrinsic parameters A, B, and S of the membrane [40]. These parameters vary depending on the structure of the membrane and the types of solutes utilized.

The membrane structural parameter, S, is key when determining the performance of FO membrane and performance. The ICP within the support layer of the membrane can be determined using the S parameter as it is a constant which is dependent on the concentrations of the DS and FS. S can be denoted as:

$$S = \left(\frac{A\pi_{ds} + B}{A\pi_{fs} + B + J_w} \right) \frac{D}{J_w} \tag{1}$$

Where A is the water permeability (L m^{-2} h^{-1} bar^{-1},), B is the salt permeability (L m^{-2} h^{-1}), D is the salt diffusion coefficient (m^2 h^{-1}), J_w is the water permeation flux (L m^{-2} h^{-1} or LMH), π_{ds} is the bulk osmotic pressure of the DS (bar), and π_{fs} is the bulk osmotic pressure of the FS (bar). The equation for water permeability, A, can be noted as:

$$A = \frac{J_w}{\Delta P} \tag{2}$$

Where ΔP is the transmembrane pressure. B, the salt permeability can be denoted as [15, 41]:

$$B = \left(\frac{1}{R} - 1 \right) J_w \tag{3}$$

Where R is the salt rejection, which can be denoted by:

$$R = \frac{C_f - C_p}{C_f} \cdot 100\% \tag{4}$$

Where C_f is the salt concentration of the FS (M) and C_p is the concentration of the DS (M). J_w, the water permeation flux, is denoted as:

$$J_w = \frac{Q}{A_m \cdot \Delta t} \tag{5}$$

Where A_m is the effective membrane area (m^2), and Q is the volume of the water permeate (L) that is collected over an elapsed time of Δt (h). In AL-DS mode, also known as PRO mode, J_w can be denoted as [15, 42]:

$$J_w = \frac{\frac{1}{K}\ln\left(A\pi_{ds} + B - J_w\right)}{A\pi_{fs} + B} \tag{6}$$

Where K is the solute resistivity (d m^{-1}), and A is the water permeability. In AL-FS mode, also known as FO mode, J_w can be denoted as [15, 42]:

$$J_w = \frac{\frac{1}{K}\ln\left(A\pi_{ds} + B\right)}{A\pi_{fs} + B + J_w} \tag{7}$$

The solute resistivity (m), K, can be denoted as [15, 42]:

$$K = \frac{t_s\tau}{D\varepsilon} = \frac{S}{D} \tag{8}$$

Where t_s is the thickness of the support layer, ε is the porosity of the membrane, and τ is the tortuosity of the membrane. The reverse salt flux (g m^{-2} h^{-1} or gMH), J_s, can be denoted as [24, 41]:

$$J_s = \frac{\Delta C \cdot \Delta V}{\Delta t \cdot A_m} = \frac{C_t V_t - C_0 V_0}{\Delta t \cdot A_m} = B \cdot \Delta C \tag{9}$$

Where ΔC is the difference in the salt concentration of the FS across the membrane (M) and ΔV is the change of the FS volume.

The parameters can be measured and calculated to exemplify the proficiency of a membrane. There are used widely in membrane trials testing how a variable of the FO membrane may affect the membrane's efficiency.

The membrane's structure begins within its layers and what these layers consist of. As mentioned earlier, FO membranes are mainly porous asymmetric membranes which consist of a dense active layer, with a thickness from 0.1 to 1 μm, that is supported by a highly porous support layer, with a thickness from 100 to 200 μm [18, 25]. The thickness of and the pore size, from 0.4 to 1 nm, the active layer determines the chemical structure of the membrane.

There are many ways to directly measure the intrinsic structural parameter of the membrane. These can be broken into two groups, imaging and analytical characterization techniques. The main imaging characterization techniques use to directly measure the intrinsic structural parameter of the membrane are x-ray tomography, confocal laser scanning microscopy, and scanning electron microscopy. The main analytical characterization techniques used to directly measure the intrinsic structural parameter of the membrane are intrusion and extrusion porosimetry, gravimetric analysis of porosity, and electrochemical impedance

spectroscopy. Once these properties of the membrane have been measured or calculated, they can be used to predict other parameters of occurrences. S of the support layer that cannot be lower than the thickness of the support layer can be used to predict ICP. A high value for S indicates a high amount of ICP and thus a low amount of water flux. Therefore, an ideal membrane would have a low S value. Membrane permeance can also be used to indicate water flux. Low permeance, less than 0.5 L m^{-2} h^{-1} bar^{-1}, results in little to no high flux meaning a less ideal membrane. The ideal water permeability of an FO membrane is at least 1 L m^{-2} h^{-1} bar^{-1}. Thus, an ideal membrane would need a medium, 0.5 t0 2.0 L m^{-2} h^{-1} bar^{-1}, or higher than 2.0 L m^{-2} h^{-1} bar^{-1}, value of A [29].

The active layer has several core characteristics, molecular weight, roughness of its surface, and thickness, and determines the separation properties of the membrane. As mentioned earlier, FO membranes require a highly rejecting active layer to preserve a high driving force from its osmotic pressure difference. There are three main methods to fabricate the active layer:

- Interfacial polymerization (IP)
- Layer-by-layer assembly (LbL)
- Conventional phase inversion
- Surface blending & grafting

The IP method is used to fabricate a thin polyamide layer as the active layer. The TFC membranes fabricated via IP have shown high salt rejection and high water permeability capabilities [43]. The IP methodology creates a thin film by dissolving two highly reactive monomers into two immiscible liquids, which in turn causes a reaction [43, 44]. The monomers used in IP are usually nucleophile reactants (typically multi-functional amines) and electrophile reactants (typically multi-functional acid chlorides) [43, 45]. The most popular monomers used are m-phenylenediamine (MPD) and trimesoyl chloride (TMC) [45]. The characteristics of TFC active layers via IP are determined by choice of the monomer, reactivity, diffusivity, concentration, and solubility. The characteristics of the support layer, pore size, porosity, roughness, and hydrophilicity play an important role when considering the characteristics of TFC layer [45]. The ideal support layer surface for IP is hydrophilic and has a pore size from 1 to 100 nm. Additionally, TFC membranes can be improved through the incorporation of carbon nanotubes or aquaporins. The polyamide layer forming through IP is also influenced heavily by the IP's experimental conditions. IP requires high experimental sensitivity and thus is complex to reproduce.

The LbL assembly method is where a thin selective film is formed by placing polyelectrolytes (PE), an oppositely charged layer, on top of a charged layer. The LbL method allows for more control precision with layers, ultra-thin defect-free layers, and various types of PE to be incorporated [29]. Until like IP, LbL assembly is straightforward and does not require complex instruments. LbL membranes can also increase water permeability, thermal stability, and high solvent resistance through the creation of an ultra-thin defect-free layer [46]. LbL membranes have limited options for DSs due to their low rejection of small draw solutes, specifically NaCl [29].

Phase inversion is a method commonly used to fabricate asymmetric membranes. This method is used in the fabrication of most commercially sold membranes. This process begins with liquid–liquid demixing to create a polymer

solution. The solution is then altered thermodynamically to create an active layer that is in turn cross-linked to the support layer. The demixing portion of the phase inversion process can be done through several methods: immersion precipitation, controlled evaporation, thermal precipitation, and precipitation from the vapor phase [47].

Surface blending is a method where materials are added to alter and improve the physical and chemical properties of the membrane. Nanoparticles and nanofibers have been incorporated to enhance and strengthen the membrane [48]. Surface grafting is where inorganic nanoparticles or organic functional groups are grafted onto the surface of the support layer. The several methods utilized for surface grafting: plasma discharge, UV irradiation, and ozone [49]. Surface grafting allows for more integration into the polymer's structure and nanocomposite membrane stability [29]. Another method used for asymmetric membranes is phase separation. Phase separation is used for hollow fiber (HF) membrane fabrication. Hollow fiber membrane fabrication is where the active and support layers are fabricated, via spinning, concurrently.

The support layer is usually fabricated using phase inversion, typically immersion precipitation, as discussed earlier. The support layer determines the water flux due to ICP. Because FO membranes do not require external pressure, they require less mechanical strength. Though less mechanical strength is required, the mechanical strength of the membrane is still essential, just less dense. A central part in the fabrication of the support layer is the polymer used in the phase inversion, the composition of the polymer solution, and the coagulation bath [29].

There are three FO membranes that are most commonly used: TFC-FO, asymmetric cellulose triacetate (CTA), and modified thin-film composite (TFC-RO) membranes [27, 50–52]. TFC-FO membranes are currently at the forefront of FO membrane research. The common methods in these studies are IP for the fabrication of the active layer, phase inversion for the support layer (electrospinning, vacuum filtration, immersion precipitation), membrane coating, and the integration of nanoparticles and nanofibers into the membrane [53–59]. Materials commonly used in the processes are N, N-dimethylformamide (DMF) within the support layer, polyamide for the active layer, and NaCl as DS. These vary with various other materials included. There is a myriad of nanoparticles infused into the support layers of the membranes. In one recent study, polyacrylonitrile (PAN) was infused into the support layer of a TFC-FO membrane with PA via IP for the active layer and NaCl as DS [58]. This membrane was able to achieve a lower reverse salt flux, mitigated ICP, and improve its water flux. As of recent, GO is another nanomaterial imbued in TFC-FO membranes. The hydrophilic nature of GO aids in the exchange between FS and DS. A comparison of all GO FO membranes was concluded by Wu et al., where all the data GO membranes were collected and analyzed. All of the membranes in this study showed the capability to improve water permeability, salt rejection efficiency, support mechanical strength, and provided remarkable antifouling properties [60]. Additionally, HF FO membranes are also considered due to their sizable surface area compared to their volume [61]. These membranes have shown reduce ICP and have high porosity and a strong pore structure [62]. The use of nanomaterials within the support layer of various types of FO membranes has with been proven to strengthen the membrane's structure, increase water flux, and reduce ICP [63]. Other materials are being incorporated into FO membranes, such as aquaporin (AQP). These membranes are biometric membranes and standout for their high water permeability [64]. Recent studies on LbL FO membranes have also shown high water permeation flux of 100 LMH, but use 3 M $MgCl_2$ as DS, due to rejection small draw solutes.

4.1 Draw solutions for FO

The DS chosen for the FO process is integral to the overall performance of the membrane. FO membranes are driven by osmotic pressure, so if the incorrect DS is chosen, there may not be enough osmotic pressure. An ideal DS would be stable, inexpensive, and accessible, provide high osmotic pressure, offer efficient recovery methods, have low toxicity, and deliver high water flux. DS can be separated into categories [63]:

- Inorganic salts
- Organic polymers
- Magnetic nanoparticles (MNP)
- Hydrogels
- Ionic liquids

Inorganic salts as DSs have been extensively researched and tested. The most commonly used inorganic salt DSs are NaCl, $NaNO_3$, KCl, KNO_3, NH_4Cl, NH_4NO_3, $CaCl_2$, and $Ca(NO_3)_2$ [35, 65]. Sodium chloride is the most commonly used DS since it is inexpensive, accessible, very soluble, and allows for high osmotic pressure [66]. Organic polymer DSs, such as glucose and fructose, do not require regeneration as the diluted DS created is typically drinkable or dischargeable. These DSs are not typically used in PW treatment. MNPs are a DS that has become inclined to many researchers due to their high recovery rate and osmotic pressure [63]. These DSs are created by using a polyol and thermal decomposition process to create hydrophilic magnetic nanoparticles [67]. The reverse salt flux of FO processes using MNPs as their draw solute is nearly nonexistent. The complication of MNP DSs is their cost. These DS are not only preparation intensive but also cost-intensive. MNP DS also has a propensity for fouling and lower water flux. Additionally, they decompose easily and can lead to the loss of draw solutes and the contamination of the newly recovered freshwater [67]. Hydrogels and ionic liquids are relatively recent additions to the studies on DSs. Hydrogels can provide high osmotic pressure, depending on the ionic group of the hydrogel polymer.

An essential part of the selection of DS is the regeneration method that is needed. These methods are thermal separation, membrane separation, thermal/membrane hybrid separation, stimuli-responsive separation, and chemical precipitation. If these methods are not utilized or required, the dilute DS is either disposed of or useable for drinking, fertilization, etc. [16]. In the case of PW treatment, the feed solution is the PW itself. Thus, a DS is appropriate for the FO membrane process and application in question. DS must provide a strong driving force for mass transport and the energy consumption associated with reconcentrating the draw solution for continuous FO operation [50].

4.2 FO membrane fouling

Fouling occurs when particles/contaminants are accumulated or absorbed by the membrane. This phenomenon can negatively affect the permeability, selectivity, and lifespan of the membrane, thus increasing the maintenance requirements and the cost. There are the main factors that affect membrane fouling: the properties of FS and the operating conditions. Membrane foulants can be separated into

three categories: inorganic, organic, and biological. Fouling in membranes treating PW is specifically caused by the accumulation or absorption of oil droplets or solutes on the surface and the internal pore structure of the membrane. Fouling caused by PW can become extremely complex, with the possibility of all three foulants happening simultaneously. In a recent study, thin-film HF membranes were used to treat PW from unconventional oil and gas fields [68]. Several trials using this membrane, using two different PW samples and in the FO and PRO modes, were completed and compared. The results showed that when the membrane was in the PRO mode, it had a high propensity for fouling. It also showed that the faster the crossflow velocity, the more likely foulant build-up would occur. Comparatively, CTA membranes are more resistant to fouling than TFC membranes when treating PW [17]. Because fouling is generally unavoidable, strategies for cleaning the membranes and fouling mitigation must be employed. The most common methods employed are pretreatment, physical cleansing, chemical cleansing, and incorporating nanomaterials into the membrane, as discussed earlier. Most membranes require water flushing followed by chemical cleansing [17, 63].

4.3 FO membrane hybrid & integrated processes

To further improve efficiency, FO membranes that are useful on their own can be paired with other treatment methods. A widely studied hybrid FO process is FO integrated with RO. A FO-RO hybrid system is where one process acts as the treatment method and the other acts as the posttreatment method. The combination of the two membrane processes leads to over a 70% increase in potential water recovery [39]. This same method can be done with NF, MF, MD, OED, MED, MD, and ED. FO-NF is a more economically feasible hybrid method than FO-RO [69]. ED-FO hybrid processes, also known as ED-FO renewable energy desalination (EDFORD), differ from the other hybrid approaches in the matter that the system is powered by solar energy. Comparatively, the cost of this process was higher than the traditional FO membrane processes [15]. All hybrid FO membrane processes are similarly effective in treating FS.

5. Environmental impacts and economic analysis

Water is an invaluable resource vital for society and humanity to survive, grow, and thrive. From clothes, food, and personal hygiene to transportation, energy, and sanitation, these means are crucial to day-to-day life, and water is needed for each of these processes to be executed successfully. Energy production uses large amounts of water and makes up 75% of all industrial water usage [70]. With water and energy demand on the rise and 39% of the population globally being unable to access water that has been adequately treated, other methods of water treatment must be considered. This study has focused on water treatment as it applies to PW. The management and treatment of PW from oil and gas sites are costly. Many materials and resources go into the production of oil and gas, and it creates many environmental impacts. The various methods utilized to treat PW were discussed earlier. These methods, though plentiful, can be quite costly and labor-intensive. This is why new methods are being explored.

When considering a product or service, it is imperative to consider the impacts the product or service may have, environmentally and economically. One way to do this is to perform a life cycle assessment (LCA). Most LCAs include the following elements [71]:

- Extraction and preparation of raw materials
- Manufacturing
- Distribution
- Usage
- Repair, upgrade, and maintenance
- Final disposal or recycling

These elements consider the various impacts and effects that the product or service may have on the environment or the economy.

5.1 Life cycle assessment of FO for PW treatment

Life cycle assessment is a method used to evaluate and estimate the environmental impacts of a product or service. It considers the phases that the products may go through and the impacts that coincide with them. The goal of LCA is to compile all important information regarding the materials, energy, and environmental inputs and outputs, to interpret the results of the assessment, and to utilize them for application. The LCA method has four steps:

- Definition of scope & goal
- Analysis of inventory
- Assessment of impact
- Interpretation

The first three steps are done in succession, with the fourth being an assessment of the first three. The first step, defining the scope & goal, depends on the context and circumstances of the product or service. In this case, the product/service is FO membranes as they relate to PW treatment.

The following step is the analysis of inventory, where an account of the input and output completed. This account includes the materials and energy consumed and the emissions and waste generated during the life cycle of the product. Once this detailed account of the inputs and outputs is completed, it is labeled as the life cycle inventory (LCI). The next step is to assess the impact the product/service may have on the environment. There are four steps to this assessment, though only two are required. The first is classification, where the inputs and outputs analyzed beforehand are placed into categories defined in the first LCA step. The next is characterization, where the impact of each induvial category is evaluated. Subsequently to characterization is normalization, where the results of the previous are normalized using a reference factor to clarify and simplify the results. Lastly is weighting, where all indicators and results are translated to a singular index or score [71]. The last step of the LCA is interpretation, which is the finalization of any data, results, assessment, or analysis made during the three previous steps. This step aims to for a clear presentation of the entire LCA, giving clear and concise conclusions and assessments of the scope and goal of the product/service.

Martins et al. performed an LCA on various membrane processes. They found that there is a limited amount of materials on the application of membrane processes [71]. To complete the LCA method explicitly, more studies on membrane processes need to be completed. They suggested that these future studies include more details on the manufacture and preparation, maintenance, disposal, recycling, and sustainability of these membranes [71]. There are a limited number of studies discussing the social and economic impacts of membrane processes. The studies that include a discussion on these impacts do not coincide with the LCA method. The study does follow the LCA steps and uses the studies on the membranes that are available. The studies available were mostly membrane processes adopted for domestic consumption. Most of the water sources being treated were non-industrial, and were either freshwater, groundwater, or wastewater. This is not a broad enough spectrum to complete an LCA of the membranes, especially in regard to PW.

Caballero et al. completed utilizing the LCA method, though with different indicators and scales. They found that multi-effect evaporation with mechanical vapor recompression (MEE-MVR), was the best alternative to standard wastewater treatment in regards to the treatment of PW, with an environmental impact 21.9% lower than the single-effect evaporation with mechanical vapor recompression (SEE-MVR) technology [72]. The categories of their LCI that were most affected by the PW treatment were climate change and fossil depletion due to the use of electricity, stem, and evaporation technologies, depending on the method. Coday et al. also completed an LCA of PW, but with a much tighter scope. They chose to focus on Haynesville shale pit water [73]. Their study tested three different treatment methods and completed an LCA on each. These were compared to other treatment methods. The first method did not employ membrane technology, the second employed CTA FO, TFC RO, and TFC NF membranes with a NaCl DS, and the third employing osmotic dilution and the same CTA FO membrane from the second method. This study found the second scenario had the most negative impact on the environment [73].

5.2 Economic analysis of FO for PW treatment

Life cycle cost analysis (LCCA) is a method used to estimate the cost of a product or service for conception to disposal. Coday et al. also performed a cost analysis of the studies done on the Haynesville shale pit water. They concluded that employing FO membranes into treatment could be more cost-efficient when compared to the current disposal practice at Haynesville. Reuse and recycling of the water could benefit their economy by up to 60% [73]. This sentiment is echoed in Echchelh et al.'s analysis of the reuse/recycling of PW versus the current disposal method. Their study found that the cost of reusing the PW instead of disposing of it was much lower [74]. The main part of this cost variance is that the PW has to be transported and discharge, which can both be cost-intensive depending on the location of the oil or gas field. This money can instead be used to treat the PW, and the treated PW can be utilized for other purposes, such as agricultural irrigation. Other studies modeled the reused of PW in an oil/gas field, though they did not include an assessment in regards to FO membranes or the reuse of PW [72]. Another study did a cost analysis of three hybrid FO systems: FO-RO, FO-NF90, and FO-NF270 [69]. They compared the total water, chemical cleaning, DS regeneration, and overall annual cost of each FO system. All three systems had a higher overall cost compared to a seawater RO system, 37.5% higher on average [69]. This increase in cost was caused by the number of FO membrane modules required to treat the water. Though there was not a large price variation from system to system, price variation could be seen

clearly in the DS used for each system. $MgCl_2$ systems were presented with the highest annual cost; thus, due to all the systems having similar annual cost, $MgCl_2$ is the cause of the spike in cost. Earlier, the importance of DS selection was discussed. This study echoes that sentiment. The cost of this DS resulted in a much higher annual cost, 67.0% higher than the system using NaCl as their DS [69].

6. Conclusion and future perspectives

Produced water treatment is a complex and costly process. This is due to the many characteristics and contaminants in PW. Current methods used to treat PW tend to be complex, costly, and harmful to the environment. New, more efficient methods are being considered. This study focuses on the application of FO membrane processes to treat PW. As discussed, FO has a myriad of characteristics and parameters that can be tailored to meet the requirements of PW to be treated. Though these membranes have an abundance of potential, there are still challenges these membranes must face. FO membranes are still being studied and tested. Most novel FO membranes discussed in this study are not able to be applied on a larger scale yet. There is also still the issue of finding a DS for PW water treatment that does not cost and maintenance intensive. FO membranes can also be integrated and combined with other membrane processes. These methods are currently more applicable due to these hybrid processes not requiring regeneration and having considerably lower fouling propensity. Fouling can also be mitigated by increasing or including a pretreatment process. There are currently not enough studies and applications of FO membranes in PW facilities and processes, making it complicated to perform a complete LCA and/or LCCA.

In the future, more studies where FO membranes and hybrid FO membrane systems are applied and tested using various DS should be completed. Furthermore, more of these studies should discuss the cost and environmental analysis of the membrane create. Currently, most studies include most of the formed and little to none of the latter. It's important to take into consideration not just how efficient a new treatment method is, but if efficiency is worth the cost in the long run. Currently, the LCA and LCCA done on membrane processes do show that FO membrane processes, hybrid, integrated, or not, are currently a feasible option on a large scale. Forward osmosis membranes can be competitive and applied widely but require additional research in membrane fabrication, better DS, fouling mitigation, and cleansing and application methods.

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